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Interscale Turbulence-Chemistry Dynamics with Reduced Basis Representations

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14. ABSTRACT Key results of the project include the development of a new periodization procedure to remove nonphysical content from bounded Fourier Transforms (FTF) of non-periodic signals [1] (Appendix A), identification of Fourier-physical space coherent structures in flame-vortex interactions relevant to turbulent premixed flames [1] (AppendixA), identification of dynamically-dominant inter-scale couplings in the advection terms [2] (Appendix B) and chemical reaction rates [3] (Appendix C), assessment of disparities in estimating the filtered chemical reaction rates using LES-filtered thermochemical quantities [4] (Appendix D), and quantification of structures of local subfilter scale species concentrations surrounding flame fronts in turbulent premixed flames [5] (Appendix E). Results from each of these studies are summarized next. Because our study centers on scale-based decomposition of the key dynamic variables within the large-eddy simulation framework, decompositions such as proper orthogonal decomposition and dynamic mode decomposition are not useful for our needs. We choose the multidimensional Fourier decomposition for two primary reasons. Firstly, the Fourier transform (FTF) is useful to decompose multidimensional multiscale analytic functions into scale-based series of complex exponentials that are mathematically convenient for scale-based analysis in elemental form. More importantly, however, the Fourier transform produces the most elemental description of the advective and chemical nonlinearities in the equations of motion as linear sums of nonlinear terms, each of which represents an elemental interaction among a few Fourier modes. These mathematically elegant elemental forms within the Fourier spectral description of nonlinear interscale interactions may be rearranged in order to down-select a small percentage of the most dominant nonlinear interscale couplings that contribute to the evolution of resolved-scale momentum, thermal energy and species concentration predicted in LES of flame-turbulence interactions. These are the dominant dynamical interactions that underlie the modeled terms in LES of premixed turbulent combustion.					
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Inter-scale Turbulence-Chemistry Dynamics with Reduced Basis Representations

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1. Objectives and Approach

1.1. Objectives

The large-eddy simulation (LES) framework centers on decomposition in scale; LES modeling centers on the interactions between larger “resolved-scale” (RS) and smaller “subfilter-scale” (SFS) fluctuations in all dependent variables. When applied to premixed turbulent combustion, the reality of (practical) LES is that there necessarily exists key SFS chemical kinetics and thermo-mechanical dynamics that impact RS evolution but cannot be supported by the grid. Thus, unlike LES of incompressible turbulence, where only momentum dynamics are involved, LES modeling of strongly density-varying turbulent flows in the presence of chemical reaction and heat release must characterize key impacts of SFS fluctuations on the evolution of RS momentum, thermal energy, and concentrations of many chemically-reacting species. In particular, SFS modeling is required for advective and chemical nonlinearities that couple the unresolvable evolutions of SFS variables with the evolution of RS variables supported by the effective grid. (“Effective” implies the existence of additional coarsening of the geometric grid from implicit filtering due to dissipative content in the SFS models and numerical algorithm.)

A central aim of our research program is to determine the key nonlinear RS-SFS couplings central to the evolution of coherent features of RS momentum, energy and species concentration in flame-turbulence interactions within premixed turbulent combustion. The “dynamically dominant” SFS structure is used to design mathematical forms for application within the contribution to the RS contributions to the advective and chemical nonlinearities from the interaction between SFS and RS fluctuations in momentum, thermal energy and species concentration. As part of this process we aim to ascertain the relationships between coherent structure in physical space and coherent structure in Fourier space with data from reduced physics simulations that are designed to incorporate key dynamics from flame-turbulence interactions, ubiquitous in premixed turbulent combustion. We apply this knowledge in our analyses of key features of the nonlinear RS-SFS couplings that are central to the evolution of coherent features of RS momentum, energy and species concentration.

In summary, the current research program integrates scale space and physical space to determine the characteristics of the dominant non-linear interscale interactions to new modeling strategies for LES of turbulence-flame interactions, the key dynamical feature of premixed turbulent combustion. From this exploration we determine a set of mathematical forms that estimate the SFS content contributing dominantly to key advective and chemical nonlinearities in the dynamical evolution of RS quantities in LES of flame-turbulence interactions. These deduced flame-turbulence-specific mathematical forms shall be referred to as generalized “basis functions” that, although neither orthogonal nor complete, capture key ubiquitous impacts of modeled SFS content on RS evolution of key dynamical variables and heat release.

The specific objectives of the research are three-fold. First, the kinematic physical-Fourier space structure underlying turbulence-flame interscale dynamical interactions are quantified in well-resolved flame-turbulence interactions within the classification of scale content into resolved and SFS fluctuations within the LES framework. Secondly, the dynamically dominant SFS content associated with both the advective and the chemical nonlinearities within the dynamical system of momentum, energy and species concentration are identified. Thirdly, from the previous analyses, classes of mathematical representation of dynamically-dominant SFS content associated with the different classes of non-linearity within the equations of motion are developed in the form of basis functions which are identified and evaluated for application to LES of flame-turbulence interactions.

1.2. Approach

We adopt the following five-stage research strategy, as illustrated in Fig. 1:

- Stage 1: Highly resolved first-principles-based DNS datasets of laminar vortex-flame interactions and turbulent premixed flames are developed for subsequent analysis. The simulations are designed to capture the interaction between vortex trains or quasi-homogeneous turbulence and a flame quasi-1D-in-the-mean flame brush, with an order of magnitude separation in scale between the scale of the flame and the vortex scale or integral scale of the turbulence entering the reaction zone.
- Stage 2: To identify the interscale couplings relevant to SFS modeling in large-eddy simulation, the scale content of the DNS data is identified and related to the physical space description in the DNS. We apply 2D or 3D FTF as a mathematically convenient and well understood representation of scale space with that important property that key second and third-order nonlinearities in the equations of motion for momentum, energy and species concentration reduce to linear sums of elemental nonlinearities, a form amenable to the extraction of the dynamically-dominant contributions to the nonlinearities.
- Stage 3: Before analyzing the interscale interactions for application to LES, the scales resolvable by an LES grid are identified. In this stage of the research, the RS vs. SFS fluctuations are objectively identified by requiring that at least 85% of the variance of the momentum and energy fields be captured within the resolved-scale motions, consistent with the fundamental LES framework. Here the key distinctions between the RS-SFS non-linear interactions obtained in the presence of combustion are emphasized in contrast with LES of non-reacting turbulent flows.
- Stage 4: Following the identification of RS and SFS content, the next issue is the extent to which SFS content in different regions of Fourier space contribute to the resolved non-linear interactions and the extent to which sub-regions with the totality of SFS fluctuations that dominantly contribute to the non-linear interactions. These details in Fourier space determine the level of detail required to capture SFS-RS nonlinear couplings sufficiently well to make measurable improvement in the evolution of RS variables. This research stage identifies what is the SFS content that contributes dominantly to resolved advective and chemical non-linearities.
- Stage 5: In this last stage, we aim to develop mathematical models for the forms representing the dynamically dominant SFS fluctuations as identified in stage 3. From these mathematical models, a set of basis functions for applications to new SFS strategies within the LES framework is formulated. Several aspects with regards to the structure, design and placement of such functions are addressed, and the effectiveness of the method in improving the model for the nonlinear terms in the governing equations underlying LES of flame-turbulence interactions is assessed.

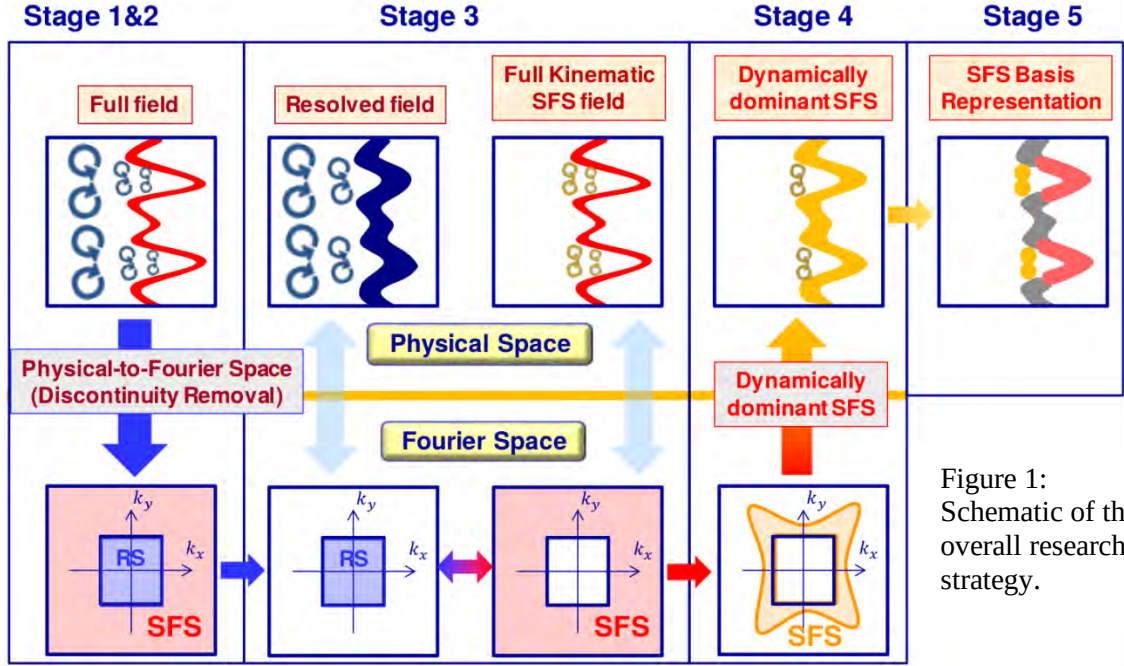


Figure 1:
Schematic of the
overall research
strategy.

2. Summary of accomplishments

Key results of the project include the development of a new periodization procedure to remove nonphysical content from bounded Fourier Transforms (FTF) of non-periodic signals [1] (Appendix A), identification of Fourier-physical space coherent structures in flame-vortex interactions relevant to turbulent premixed flames [1] (Appendix A), identification of dynamically-dominant inter-scale couplings in the advection terms [2] (Appendix B) and chemical reaction rates [3] (Appendix C), assessment of disparities in estimating the filtered chemical reaction rates using LES-filtered thermochemical quantities [4] (Appendix D), and quantification of structures of local subfilter scale species concentrations surrounding flame fronts in turbulent premixed flames [5] (Appendix E). Results from each of these studies are summarized next.

2.1. The application of the Fourier transform (FTF) and a new periodization procedure for FTF of non-periodic signals

Because our study centers on scale-based decomposition of the key dynamic variables within the large-eddy simulation framework, decompositions such as proper orthogonal decomposition and dynamic mode decomposition are not useful for our needs. We choose the multidimensional Fourier decomposition for two primary reasons. Firstly, the Fourier transform (FTF) is useful to decompose multidimensional multiscale analytic functions into scale-based series of complex exponentials that are mathematically convenient for scale-based analysis in elemental form. More importantly, however, the Fourier transform produces the most elemental description of the advective and chemical nonlinearities in the equations of motion as linear sums of nonlinear terms, each of which represents an elemental interaction among a few Fourier modes. These mathematically elegant elemental forms within the Fourier spectral description of nonlinear interscale interactions may be rearranged in order to down-select a small percentage of the most dominant nonlinear interscale couplings that contribute to the evolution of resolved-scale momentum, thermal energy and species concentration predicted in LES of flame-turbulence interactions. These are the dominant dynamical interactions that underlie the modeled terms in LES of premixed turbulent combustion.

However, to develop Fourier-based analysis of flame-turbulence interactions, we must recognize and eliminate spurious spectral content that is introduced when the FTF is applied to a signal that is nonperiodic over a bounded domain. This is because the FTF of a bounded nonperiodic continuous analytic signal produces periodic extensions of the signal in physical space with discontinuities at the boundaries. As a global basis, the corresponding Fourier

coefficients include contributions from these boundary discontinuities that complicates scale-based analysis: each boundary discontinuity creates Fourier coefficients with power law decay extending from lowest to highest wavenumber in the direction of the discontinuity. Consequently, when the highest physical gradients are resolved by the grid, the power-law artifact from the boundary discontinuity dominates the energy content of the physical signal at high wavenumbers. Several "periodization" procedures have been proposed in the literature to overcome or minimize the impacts of this unwanted spectral content ("pollution") from boundary discontinuities that overlaps the signal of interest in Fourier space). However, whereas these methods do retain spectral accuracy while removing spurious scale content by fully periodizing the signal, they also introduce new nonphysical Fourier content that overlaps with that of the original signal, making these approaches unsuitable for our applications.

We have therefore developed a "pollution removal" methodology that specifically treats the tradeoff between full periodization and alteration of the signal in regions of interest. Ours is a semi-empirical approach that removes pollution from boundary discontinuities without creating additional spurious content that overlaps the signal in regions of interest in both Fourier and physical space. This is done through user-based empiricism introduced to balance between the need to modify the signal near the boundaries for periodization with the extent to which the signal is modified away from the boundaries. We have extended our method to higher order discontinuities and multidimensional signals.

The details of our method can be found in an AIAA paper (sect. 4.1, #7: Ref. [6]), in the thesis of Paulo Paes (sect. 4.2, #1) and, most completely, in a recent paper that will soon appear in *AIP Advances* (sect. 4.1, #1: Ref. [1]). In essence, we subtract the Fourier coefficients of a sawtooth function with the same C^0 discontinuity as in the nonperiodic direction of the signal. However, the sawtooth function has been high-pass filtered to localize signal modifications to the boundaries while minimizing modifications to the signal in the defined region of interest. We propose a method that automatically balances between: (1) minimization of changes to the signal in the region of interest, and (2) minimizing the spurious spectral content from the boundary discontinuity. The user chooses a parameter that can shift the periodized signal towards (1) or (2). However, we show that with complete periodization the signal is maximally modified in our region of interest. Thus, ours is a "partial" periodization method, whereby a very small amount of residual nonperiodicity is allowed so as to minimally change the signal. This discontinuity pollution removal procedure has potential applications that extend well beyond multidimensional Fourier analysis of premixed turbulent combustion DNS.

2.2. Fourier-Physical Space Coherent Structure in Flame-Vortex Interactions

Having developed a viable discontinuity pollution removal method, we study scale-dependent coherent structure simultaneously in physical space and in Fourier scale space. A central aim of the current study is to determine the relationships among key coherent elements in momentum, thermal energy and species concentration from turbulence-flame interactions in terms of the characteristic scales of the coherent elements and their spatial inter-relationships concurrently in physical space and Fourier space. The analysis provides understanding of SFS vs. RS coherent structure in relationship to the effective LES filter cutoff within the Fourier description of scale, as determined by effective grid. This understanding will inform future analyses and a new modeling strategy for the RS-SFS contributions to the nonlinear terms in LES of flame-turbulence interactions centered on the development physically meaningful mathematical forms for scale-dependent SFS variables.

However, the three-dimensional chaotic forcings of a thin flame by the full range of turbulence eddy scales creates highly complex multiscale flame distortions in response to the wide ranges of eddy scales and strengths relative to the flame scale and flame strength local to a flame-eddy interaction. Spatial coherence resulting from these interactions is correspondingly highly complex, with coherent structure created over wide ranges of scale by the dynamics associated with the distorting flame. This complexity in multi-scale response makes it difficult to dissect the Fourier-physical space relationships created by groupings of eddies at similar scale and strength. To reduce the levels of complexity from the impacts of a full range of 3D turbulence eddying motions on flame dynamics, and to more clearly describe coherent structure physical-Fourier space relationships in context with flame topography, we have created a "reduced-physics" model that captures a key underlying element in turbulence-flame dynamics: the distortion of a flame in response to repetitive forcing by single-scale eddies of controlled scale and strength relative to a well-defined flame sheet. Specifically, we design controllable non-chaotic interactions between an initially unstretched planar flame sheet and topologically well-defined eddies modeled as trains of two-

dimensional single-scale vortices with controlled scale and strength relative to that of the flame. We argue that this flame-vortex interaction model captures essential deformations of a flame sheet in response to single-scale eddy motion. The reduction in dimensional and physical complexity produces well-defined Fourier-physical space relationships in coherent structure while retaining sufficient levels of complexity to apply the results extract key elements of turbulence-flame interactions that are local to flame-eddy interactions in premixed turbulent combustion.

With the reduced-physics flame-vortex interaction model we are able to choose the relationships between the flame scale/strength and eddy scale/strength in relationship to the interactions between a subset of eddies within the full range of turbulence-flame interactions that are ubiquitous in premixed turbulent combustion. As discussed in Sec. III B, the current study focuses on eddies with scale an order of magnitude larger than the characteristic flame scale. This ratio is chosen because turbulence eddies of order the flame scale tend not to strongly impact flame distortion²⁶, while the response to eddies larger in scale may be deduced from the current results. We argue that the current flame-eddy model characterizes local deformations of the flame sheet at the defined eddy scale with the degree of response determined by the strength of the defined single-scale eddies relative to the strength of the flame. In the complete series of studies underlying the current paper, relative eddy strengths are chosen to create “weak”, “moderate” and “strong” response in flame deformation. In the current paper we discuss in detail the “weak” response case, designed to create large repetitive distortions of the flame without pinching or the creation of pockets. The higher strength of the “moderate” case vortices was chosen to produce pinching (but no pockets), while the “strong” vortex case was designed to create repetitive pocket formation. Because the primary conclusions from this series of studies can be presented in context with “weak” case, we present detailed results for this case in the current paper. The “moderate” and “strong” cases are discussed in Sec. V in context with discussion of the application of the current analysis to 3D turbulence-flame interactions.

Our analysis is described in detail in an AIAA paper (sect. 4.1, #6: Ref. [7]), in the PhD thesis of Paulo Paes (sect. 4.2, #1), and in a paper soon to appear in *AIP Advances* (sect. 4.1, #1: Ref. [1]).

Having removed signal pollution in the nonperiodic direction, we analyzed the relationships between coherent structure in physical and Fourier space through the application of systematic localized filtering in Fourier space in context with the multidimensional Fourier representation of scale content. The aim is to find collections of Fourier coefficients connected to localized coherent structures in physical space, where “coherence” is embedded in the phase correlations that exist among individual Fourier coefficients at different scales in different directions (i.e., positions in Fourier space) underlying the physical-space representation of the coherent structure. We find that small-scale coherent structure is organized within a coherent broadband “star-like” pattern in Fourier space that extends from vortex to the flame scales. The magnitudes and directional dependences of the Fourier coefficients within the legs of the star are closely connected with the direction and magnitude of flame-normal gradients of key variables within the corrugated flame front. This observation suggests that Fourier content of flame-turbulence interactions would also be organized in coherent broadband star-like pattern over a wide range of angles due to the wide range of flame corrugations caused by turbulence. The current study shows that, in more complex multiscale turbulence-flame interactions, collections of flame segments with similar flame-normal orientation may be isolated by the systematic application of specific filtering operations in Fourier space developed in the current study. With this understanding, it is possible to analyze DNS studies to relate embedded coherence in multidimensional scale-space to specific nonlinear interactions underlying evolution in physical space.

For a consistent interpretation of our reduced-physics simulations in relationship to LES, we defined resolved scales with a 2D Fourier space filter that decomposes scale-based content of fluctuations in key variables into RS vs SFS based on the key criterion underlying LES that the spectral content of the variances of key variables is captured by the effective grid to the 80-90% level. Interestingly, our analysis shows direct correspondence between localized regions of RS Fourier space and coherent structural features in physical space at the resolved scales, implying that it is now possible to isolate the interscale couplings underlying the evolution of specific structural features. Most importantly we have identified the localized regions in 2D Fourier space that correspond to the corrugations in the flame front in the transverse direction in response to forcing by the vortex train, as well as axial gradients associated with the overall planar structure of the flame. In addition, we have shown that the elongated structures within the products that are created as eddies pass through the flame are localized in Fourier space to axial wavenumbers near $k_x = 0$ and spanwise wavenumbers that match the dominant spanwise wavenumbers k_y of the forcing eddies. We anticipate that these general groupings of Fourier modes exist in fully turbulent flame-

turbulence interactions with details dependent on the scales and strengths of the turbulence eddies that distort the flame front, making it possible to study nonlinear scale interactions associated with specific features underlying flame-turbulence dynamics in premixed turbulent combustion.

We developed two additional computational models with “moderate” and “strong” vortices, where the moderate vortices generated long neck distortions of the flame sheet and strong vortices produce pinching of necks into pockets that continue to burn within the burnt products. There were a few findings that add significantly to what has been discussed here. Primarily it was found that the stronger-vortex cases emphasize the kinematic Fourier-physical-space relationships associated with strongly corrugated flame sheets. Specifically, the variance content of flame corrugations increases relative to the vortices. In addition, the star-like pattern becomes buried within frontal contributions induced by discontinuities at the surfaces of the pockets which radiate in all directions and obscure the directions of larger-area flat portions of the primary flame sheet. Extrapolating to fully-developed turbulence, one anticipates that the star-like pattern becomes a roughly isotropic distribution of star-like patterns. In the current study we learn that individual kinematic contributions to Fourier-space structure from frontal content in the presence of the full range of turbulence scales can be extracted with the application of elliptical filters extended to 3D Fourier space similar to the application of three-dimensional wavelet filtering with physically-modeled wavelet scales.

2.3. Dynamically-dominant inter-scale couplings in the advection terms

The FTF of the advective nonlinearities in the governing equations for momentum, energy, and species concentration in turbulent combustion, for example, appear as linear sums of elemental nonlinear “triadic” interactions among three Fourier modes with wave-vectors that form a closed triangle in multidimensional Fourier space. We make use of this Fourier-spectral property to analyze the nonlinear interactions among scales of motion within turbulent-reacting flows in context with LES, where turbulence structure is separated into RS and SFS spectral content:

$$\frac{\partial \beta^r}{\partial t} = -\nabla \cdot (\beta \bar{u})^r = -\nabla \cdot (\beta^r \bar{u}^r)^r - \nabla \cdot (\beta^r \bar{u}^s + \beta^s \bar{u}^s)^r + \dots, \quad (1)$$

where $\beta(\bar{x}, t) = \beta^r(\bar{x}, t) + \beta^s(\bar{x}, t)$, $\bar{u}(\bar{x}, t) = \bar{u}^r(\bar{x}, t) + \bar{u}^s(\bar{x}, t)$. (2)

Whereas $\bar{u}(\bar{x}, t)$ is the velocity field at time t , $\beta(\bar{x}, t)$ represents momentum, enthalpy (or internal energy) or species concentration, all on a per unit volume basis. Equation (1) is the second-order advective nonlinearity in the governing equation for the effective-grid-resolved variable $\beta^r(\bar{x}, t)$ as indicated by superscript r . Superscript s refers to SFS variables that are not supported by the LES effective grid.

Given the nonexistence of SFS quantities in a LES, the second term on the RHS of eq. (1) must be modeled. These “modeled terms” represent interactions between SFS motions and RS motions in the evolution of the RS variable on the LHS. Our basic modeling strategy is to recognize that, whereas grid support does not exist for SFS variables β^s and $\bar{u}^s$ in the modeled terms, so they cannot be represented at their proper scales, they can be modeled with mathematical forms at the proper scale. The central aim of the current study is to determine canonical physical-space structure of the “dynamically dominant” contributions to the SFS variables on the RHS of Eq. (1). By “dynamically dominant” we mean the SFS contributions to β^s that contribute dominantly (i.e., most strongly) to the modeled contribution to the advective nonlinearity which, itself, is a primary contributor to the time derivative and space-time evolution of $\beta^r(\bar{x}, t)$. By “canonical” we mean we search for basic structural forms that can be approximated with basic mathematical forms (“basis functions”) that would be parameterized and inserted on the RHS of Eq. (1) as a model of the SFS contribution to the advective nonlinearity. The current study stopped short of parameterization and focused on the existence of canonical structural forms in physical space and the viability of representing these structural features with simple mathematical expressions that could be placed within the computational domain to approximate the modeled term in eq. (1) over regions of interest within the computational domain.

A key element in the current analysis is the determination of the SFS contributions to the dynamically dominant RS-SFS interactions represented by the SFS contribution to the nonlinearity. To determine dynamical dominance

we take advantage of a key property in the Fourier description: the FTF of the nonlinearity is mathematically given by the sum of elemental interactions among three Fourier modes in groupings called “triads”:

$$\left. \frac{\partial \hat{\beta}(\vec{k}^r)}{\partial t} \right|_{adv} = \sum_{\vec{p}=\vec{k}^r-\vec{q}} \hat{\beta}(\vec{p}) [\vec{k}^r \cdot \hat{u}(\vec{q})] = \sum_{\vec{p}=\vec{k}^r-\vec{q}} \hat{\Delta}(\vec{k}^r; \vec{p}, \vec{q}) \quad (3)$$

where $\hat{\beta}(\vec{k}, t)$ is the FTF of $\beta(\vec{x}, t)$ and $\hat{\beta}^r(\vec{k}^r, t)$ is the FTF of $\beta^r(\vec{x}, t)$. The triangle symbol represents a single triadic interaction. The linear sum of elemental triadic interactions is indicative of the elemental nature of the FTF, as previously discussed. We take advantage of the linear nature of the sum to develop a methodology to determine the dominant contributors to the series within numerical simulations that represent canonical flame-turbulence interactions. We refer to this methodology as “the down-select process.”

Once the down-select process is complete and the sum in eq. (3) is reduced to a partial sum that contains only the dynamically dominant contributions to the sum, the SFS content of the variable of interest is extracted and inverse-transformed to pseudo-spectral to determine and quantify the structure of the dynamically dominant SFS contribution to that variable in the SFS contribution to the advective nonlinearity. Thus, whereas Fourier space is essential to the down-select methodology, we ultimately characterize dynamically dominant SFS structure in physical space where we aim to identify and quantify canonical structure that could underlie the design of basis functions.

The down-select process involves the use of both amplitude and phase information for each term in the triadic sum. The details are described in detail in an AIAA paper (sect. 4.1, #5: Ref. [2]) and in the PhD thesis of Paulo Paes (sect. 4.2, #1). The simulations are of two types, the reduced physics simulations of flame-vortex interactions discussed in the previous section and DNS of full turbulence-flame interactions discussed in section 2.5. The detailed analysis of the of the flame-vortex simulations are described in the references above where use was made of the new discontinuity removal periodization procedure described in sect. 2.1 and the analysis of Fourier-physical space coherent structure in flame-vortex interactions described in sect. 2.2. The analysis was carried out for momentum/volume, enthalpy/volume and several key species within key reactions.

Two key important results emerged from our detailed analysis of flame-vortex interactions that we subsequently found also to be the case with our DNS of flame-turbulence interactions (sect. 2.5):

Firstly, the dynamically dominant SFS structure is found to lie near the center of the flame sheet (the reaction zone), as opposed to pervasively spread throughout the turbulent flow region. This is an important result from a modeling perspective because it implies that a zeroth order modeling strategy should be sufficient to improve predictions of key nonlinearities in the evolution of the resolved-scale variables. Localizing model improvements to the flame sheet implies a very large reduction in spatial degrees of freedom and, correspondingly, suggest that modeling strategies using this direction of research may be computationally reasonable—a key practical requirement for any advanced modeling strategy. Furthermore, the location of the flame sheet can be well approximated from predictions of resolved-scale variables, specifically key species that are generated within the reaction zone.

The second key result, also important to a viable SFS modeling strategy, is that the discovery of two (or possibly three) dominant structural forms for dynamically dominant SFS structure of key variables (momentum, energy, key species concentrations). We have labeled these forms as “single peaked,” “double peaked,” and “transitional,” where the latter implies a transition within a dominantly single peaked or double peaked forms. As discussed in the references identified above, these forms are parameterized in terms of (i) location relative to the center of the flame sheet (which we show to reasonably well quantified as the isosurface 0.5 of the resolved temperature-based progress variable), (ii) an amplitude variable, (iii) a scale, and (iv) symmetry of structure relative to reaction vs. products side of the flame sheet. We further showed that over “low curvature” parts of the flame (over 90% in the flame-turbulence interactions) these structure forms can be represented with 1D mathematical forms that, when accurately parameterized, can increase the accuracy of the advective nonlinearity by factors of 10. However, we also find that the SFS contribution to the advective nonlinearities is of order 10%, lower than was anticipated.

2.4. Dynamically-dominant inter-scale couplings in the chemical source terms

Similar to the advective nonlinearity, the FTF of the chemical nonlinearities, the source terms in the governing equations for species concentration, appear as linear sums of elemental nonlinear interactions. However, we consider third-order chemical nonlinearities where the FTF of the nonlinearity creates linear sums among four Fourier modes with wave-vectors that form a closed quadrilateral in multidimensional Fourier space. Like the advective nonlinearity, this Fourier-spectral property whereby interscale interactions are represented as linear sums over elemental nonlinear terms is key to the down-select process. However, the involvement of four Fourier modes, rather than 3, increases the complexity of the elemental terms:

$$\left. \frac{\partial \theta_i^r}{\partial t} \right|_{chem} = \sum_{j \text{ reactions}} [\kappa_j^r(T) \theta_{m(j)}^r \theta_{n(j)}^r]_i^r + \sum_{j \text{ reactions}} [\kappa^r \theta_m^r \theta_n^s + \kappa^s \theta_m^r \theta_n^s + \kappa^r \theta_m^s \theta_n^r + \kappa^s \theta_m^s \theta_n^r + \kappa^r \theta_m^s \theta_n^s + \kappa^s \theta_m^s \theta_n^s + \kappa^s \theta_m^s \theta_n^s]_{j,i}^r \quad (4)$$

where $\theta_i(\vec{x}, t) = \theta_i^r(\vec{x}, t) + \theta_i^s(\vec{x}, t)$ is mass concentration for species i , κ is the temperature-dependent Arrhenius prefactor, and the sum is over all chemical reactions that contribute to the chemical source term in the evolution equation for θ_i . Conceptually, the modeling concept is the same as with the advective nonlinearity: search for canonical forms for the dynamically dominant SFS variables that dominantly contribute to the modeled term (the second term with SFS contributions) in eq. (4).

Similarly to the advective nonlinearity, the down-select process is developed for the FTF of (4) given by a linear sum over nonlinear terms each of which represents a “quadrad”:

$$\left. \frac{\partial \hat{\theta}_i(\vec{k}^r)}{\partial t} \right|_{chem,j} = \sum_{\vec{l}=\vec{k}^r-\vec{p}-\vec{q}} \hat{\kappa}_j(\vec{p}) \hat{\theta}_{m(j)}(\vec{q}) \hat{\theta}_{n(j)}(\vec{l}) = \sum_{\vec{l}=\vec{k}^r-\vec{p}-\vec{q}} \hat{\square}_j(\vec{k}^r; \vec{p}, \vec{q}, \vec{l}), \quad (5)$$

where the square symbol represents a single quadrad. The down-select process is centered on the development of a methodology to identify the quadrad terms that contribute dominantly to the RHS of eq. (5). The details of the down-select process are described for the flame-vortex interaction model in the PhD thesis of Paulo Paes (sect. 4.3, #1). We are currently developing a manuscript on this analysis for submittal to an international journal. The down-select process for the full flame-turbulence interactions model is described in the PhD thesis of Yash Shah (sect. 4.2, #2). Due to the unfeasible level of computational expense required to calculate the huge number of quadrad terms on the RHS of eq. (5) for a full 3D turbulent flow, the down-select process was modified and an approximation introduced to allow down-select within a single the sum of quadrad terms to be approximated using two down-selects of sums over triad terms to a regrouping of the RHS of (5). Once the dynamically dominant terms are identified for key reactions, j , the SFS content of species $\hat{\theta}_{m(j)}$ and $\hat{\theta}_{n(j)}$ in these terms is extracted and inverse-transformed to physical space.

Much analysis of the dynamically dominant SFS content of key species in key reactions was develop both for the 2D reduced physics flame-vortex interaction model (PhD thesis of Paulo Paes) and for the fully turbulent 3D flame-turbulence interactions model (PhD thesis of Yash Shah). These analyses are described in an extended abstract for the 11th U.S. National Combustion Meeting in 2019 (sect. 4.3, #3) and in the PhD thesis of Paulo Paes (sect. 4.2, #1) and in a paper soon to appear in the *Physics of Fluids* (sect. 4.1, #2; Ref. [5]). The latter analysis is described in the next section.

A number of important discoveries were made in these studies. Firstly, similar to the advective nonlinearity, dynamically dominant SFS structure species concentrations are localized near the flame sheet, again implying a zeroth order modeling strategy to improve predictions of key chemical nonlinearities in the evolution of resolved-scale species concentrations. Given the complexity and number of key reactions involved in the chemical nonlinearities, the large reduction in spatial degrees of freedom is critically important to practical modeling strategies. However, we also find, similarly to the advective nonlinearity, the existence of two primary structural forms for dynamically dominant SFS structure of species concentration, single and double peaked structure parameterized by location, amplitude, scale and symmetry. Whereas we again find that these structure forms can be represented with 1D mathematical forms over 90% or more of the flame sheet, we also find that adjustments in form must be made in regions when neighboring parts of the flame sheet are within roughly 3 laminar flame

thicknesses (section 2.5). Another important difference between the chemical and advective nonlinearity is that whereas the SFS contribution to the advective nonlinearities is only of order 10%, the SFS contribution to the chemical nonlinearity typically exceeds 40%, based on our *a posteriori* analysis of flame-turbulence interactions. We conclude that our new modeling strategy should focus more on the chemical nonlinearity than on the advective nonlinearity.

2.5. Assessment of disparities in estimating the filtered chemical reaction rates

To quantify the effects of SFS thermochemical variables on the estimation of filtered chemical reaction rates in LES, we performed an *a priori* assessment of the disparities in estimating the filtered chemical reaction rates using two DNS datasets of turbulent premixed flames, as described in detail in [4] (Appendix D). DNS of turbulent premixed flames with the same turbulent Reynolds number but different Karlovitz numbers were carried out using semi-detailed finite-rate chemistry to represent different flame conditions. The sensitivities of the differences between the LES-filtered chemical reaction rates and those constructed using LES-filtered thermochemical variables (referred to as ‘disparity’ below) to reaction types, species characteristics, and flame conditions were systematically investigated. The sensitivity to reaction type was examined by quantifying the disparity in the filtered temperature-dependent reaction rate constant as a function of kinetic rate parameters. The sensitivity to species characteristics was examined by quantifying the disparity in the filtered law of mass action as a function of species chemical time scales and formation location. The sensitivity to flame conditions was examined by contrasting the results obtained from the two simulations. These disparities were compared to characterize distinct reaction/species categories with different levels of disparity in estimating the filtered chemical reaction rates in LES.

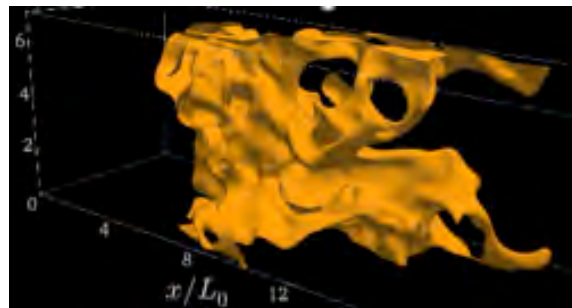


Figure 2. An iso-surface of the highly distorted flame from DNS of a turbulent premixed flame.

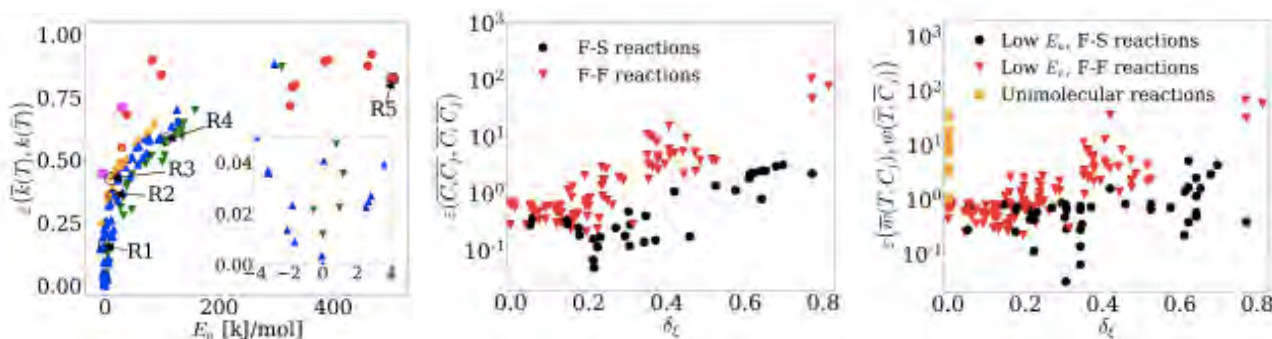


Figure 3. Normalized disparities in estimating the Arrhenius rate constants, law of mass action, and chemical reaction rates for all reactions in the chemical model used. “F” denotes a “Fast” species with chemical time scales smaller than $10 \mu\text{s}$. “S” denotes a “Slow” species with chemical time scales larger than $10 \mu\text{s}$.

It was found that the rate constants estimated using LES-filtered temperature severely under-predict the LES-filtered rate constants, especially for reactions with high activation energies, E_a (see Fig. 3a). The law of mass action estimated using LES-filtered species concentrations severely over-predicts the LES-filtered values, especially for reactions involving reactants with shorter chemical time scales present at different locations within the turbulent flame structure (see Fig. 3b). Combining the two effects, it was found that bi-molecular reactions typically involve at least one reactant with small chemical time scale (e.g., radicals), which makes the disparity in estimating the filtered chemical reaction rates generally high for these reactions. Unimolecular reactions are typically associated with large E_a , which makes the disparity even higher (see Fig. 3c). The disparity in estimating the filtered chemical reaction rates would be low only for bi-molecular reactions between reactants with large chemical time scales,

typically stable molecules, and with low E_a , however, such reactions are rare since reactions between stable molecules typically require very large E_a . These results were found independent of filter width.

2.6. Quantification of structures of local subfilter scale species concentrations

We identified the structures of SFS thermochemical quantities, such as temperature and species concentrations, important to the estimation of chemical reaction rates in LES, using *a priori* analysis of DNS of a turbulent premixed flame with semi-detailed finite-rate chemistry, as described in detail in [5] (Appendix E). The significance of these SFS quantities was quantified by computing the differences between the complete chemical reaction rates extracted from DNS and those estimated from LES-filtered variables. We defined the SFS “influence zones” as regions where such differences are large, and these zones were found to be localized around the flame only within a few laminar flame thicknesses away from the flame front (defined as the iso-surface of a temperature-based progress variable of 0.5). Within these zones, variations in the location relative to the flame front, scale, and magnitude of the SFS thermochemical quantities were quantified and the mean SFS structures were determined for a number of representative species. Two types of SFS species structures were identified: “single-banded” structures characterized by one distinct peak, and “double-banded” structures characterized by two peaks of opposite signs (see Fig. 4). Species that are produced and consumed within the flame such as CH_2O and HCO are observed to have single-banded structures and species displaying a frontal behavior such as $\text{n-C}_7\text{H}_{16}$ (fuel) and OH are found to have

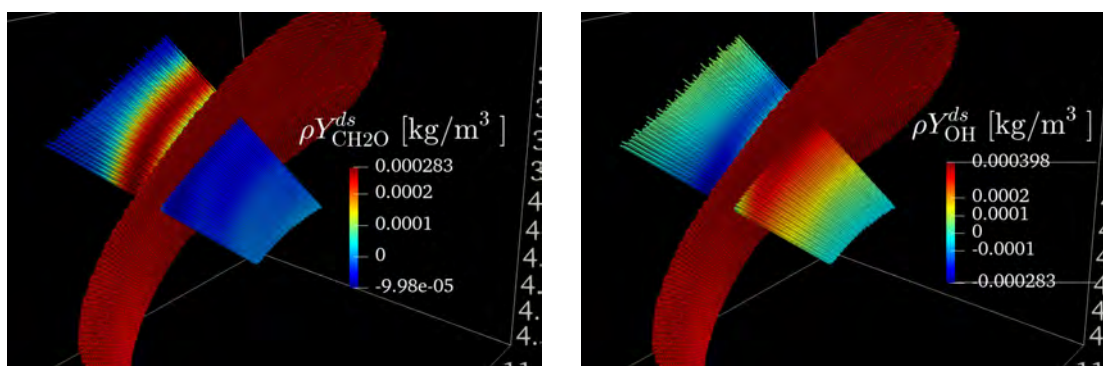


Figure 4. Iso-contours of the dynamically-dominant SFS contributions relative to a portion of the flame front (red). Left: structure of formaldehyde (CH_2O) is single-banded. Right: structure of OH is double-banded.

double-banded structures, on average. The local SFS structures surrounding the flame were found to be affected by neighboring flame-flame interactions as well as by variations in flame curvature. The effects of the flame-flame interactions were observed to be generally strong when a neighboring flame front is present within 3 laminar flame thicknesses. Beyond such distance, curvature effects are shown to be strong.

3. Summary

This research program integrated Fourier-based scale space and physical space to determine the characteristics of the dominant non-linear interscale interactions to new modeling strategies for LES of turbulence-flame interactions, which is a key dynamical feature of turbulent combustion. From this exploration, we determined a series of structural forms that can approximate the SFS content contributing dominantly to key advective and chemical nonlinearities in the dynamical evolution of RS quantities in LES of turbulent flames. These deduced structural forms can potentially be modeled by analytical functions, referred to as generalized “basis functions”, that, although neither orthogonal nor complete, capture key ubiquitous impacts of modeled SFS content on RS evolution of key dynamical variables and heat release.

Our work explores the possibility for a new LES modeling approach in which the SFS fields are approximated with analytical expressions to reconstruct the unclosed terms. This is different from most existing modeling approaches, which target the unclosed terms in LES of turbulent flames. This strategy could be advantageous since, once developed, the models for SFS quantities can be applied to both unclosed scalar flux terms and chemical reaction rates. Moreover, directly modeling these SFS quantities would, in principle, allow the LES solution to approach the DNS solution progressively with grid refinement.

4. Publications, Abstracts and Presentations

4.1. Publications

1. Paes, P.L.K., Brasseur, J.G., Xuan, Y., Shah, Y.G. 2021 Fourier-Physical Space Coherent Structure in Flame-Vortex Interactions Relevant to Flame-turbulence Interactions using a New Signal Periodization Procedure, In press, *AIP Advances*, <https://doi.org/10.1063/5.0050280>.
2. Shah, Y.G., Brasseur, J.G., Xuan, Y., 2021 On the Structure of Local Subfilter-scale Species Surrounding Flame Fronts in LES of Premixed Turbulent Combustion, *AIP Advances*, in press, *Physics of Fluids*, <https://doi.org/10.1063/5.0045870>.
3. Shah, Y.G., Brasseur, J.G., Xuan, Y. 2020 Assessment of disparities in estimating filtered chemical reaction rates in LES using DNS of turbulent premixed combustion. *Combustion Theory and Modelling*, <https://doi.org/10.1080/13647830.2020.1831616>.
4. Paes, P.L.K., Shah, Y.G., Brasseur, J.G., Xuan, Y. 2020 A Scaling Analysis for the Evolution of Small-Scale Turbulence Eddies across Premixed Flames with Implications on Distributed Combustion. *Combustion Theory and Modelling*, 24(2), 307-325, <https://doi.org/10.1080/13647830.2019.1679396>.
5. Paes, P.L.K., Brasseur, J.G., Xuan, Y. 2019a A Novel Strategy to Identify Dynamically Dominant Inter-Scale Couplings for Application to Large-Eddy Simulation of Premixed Turbulent Combustion. AIAA Paper No. 2019-1644, January 2019 SciTech Meeting, San Diego.
6. Paes, P.L.K., Brasseur, J.G., Xuan, Y. 2019b Kinematic Relationships between Physical and Fourier Space in Premixed Turbulent Combustion for Application to Large-Eddy Simulation. AIAA Paper No. 2019-2145, January 2019 SciTech Meeting, San Diego.
7. Paes, P.L.K., Xuan, Y., Shah, Y.G., Brasseur, J.G. 2019c Removing Pollution in the Fourier Space Representation of Non-Periodic Signals over Finite Domains, AIAA Paper No. 2019-0903, January 2019 SciTech Meeting, San Diego.

4.2. Ph.D. Theses

1. Paes, Paulo, 2019. *Topic: Interscale Turbulence-Chemistry Dynamics with Reduced Basis Representations*, Ph.D Thesis, Pennsylvania State University, University Park, PA, May 2019.
2. Shah, Yash, 2021. *Topic: Dominant Interscale Dynamics in Premixed Turbulent Combustion for Application to Large-Eddy Simulations*, Ph.D Thesis, Pennsylvania State University, University Park, PA, March 2021.

4.3. Abstracts

1. Brasseur, J.G., Paes, P.L.K., Xuan, Y. 2020 Dynamically-dominant Subfilter-scale content for application to LES of Turbulence-Flame Interactions in Premixed Turbulent Combustion. (abstract) 73rd Annual Meeting of the APS Division of Fluid Dynamics, <http://meetings.aps.org/Meeting/DFD20/Session/P03.10>.
2. Brasseur, J.G., Shah, Y., Paes, P.L.K., Xuan, Y. 2019, Fundamental Differences between Large-Eddy Simulation of Incompressible Turbulence vs. Premixed Turbulent Combustion, (abstract) Bull. Amer. Phys. Soc. 64 (13): <https://meetings.aps.org/Meeting/DFD19/Session/NP05.160>.
3. Paes, P.L.K., Shah, Y.G., Xuan, Y., Brasseur, J.G. 2019 Dynamically Dominant Interscale Couplings in the Nonlinear Chemical Source Terms for Species Evolution in Premixed Turbulent Combustion with Application to LES Modeling. 11th U.S. National Combustion Meeting, Pasadena, California, March 24-27, 2019.
4. Xuan, Y., Brasseur, J.G. 2018 Inter-scale turbulence-chemistry couplings in premixed turbulent combustion for large-eddy simulations. Bull. Amer. Phys. Soc. 63 (13): <http://meetings.aps.org/link/BAPS.2018.DFD.L25.1>.

5. Brasseur, J.G., Paes, P.L.K., Xuan, Y. 2018 Dominant Subfilter-Resolved Scale Flame-Turbulence Advective Nonlinearities in LES of Premixed Turbulent Combustion. (abstract) Bull. Amer. Phys. Soc. 63 (13): <http://meetings.aps.org/link/BAPS.2018.DFD.A02.8>.
6. Shah, G., Paes, P.L.K., Brasseur, J.G., Xuan, Y. 2018 Concurrent relationships between Physical and Fourier Space representation of three-dimensional flow structures for LES of premixed turbulent combustion. (abstract) Bull. Amer. Phys. Soc. 63 (13): <http://meetings.aps.org/link/BAPS.2018.DFD.A02.7>.
7. Paes, P.L.K., Brasseur, J.G., Xuan, Y. 2017 Relationships between Physical and Fourier Space for Large-Eddy Simulation of Premixed Turbulent Combustion: Multi-Dimensional Fourier Decomposition. (abstract) Bull. Amer. Phys. Soc. 62 (14): <http://meetings.aps.org/link/BAPS.2017.DFD.F2.1>.
8. Xuan, Y., Paes, P.L.K., Brasseur, J.G. 2017 Relationships between Physical and Fourier Space for Large-Eddy Simulation of Premixed Turbulence Combustion: Transition from Weak to Strong Vortices. (abstract) Bull. Amer. Phys. Soc. 62 (14): <http://meetings.aps.org/link/BAPS.2017.DFD.F2.2>.
9. Paes, P.L.K., Brasseur, J.G., Xuan, Y. 2017 Global and Local Response of Premixed Flames during Flame-Vortex Interactions under Distinct Configurations. 10th U. S. National Combustion Meeting, April 23-26, 2017, College Park, MD
10. Paes, P.L.K., Brasseur, J.G., Xuan, Y. 2016 Representation of the Essential Flame-Turbulence Dynamics using Specific Flame-Vortex Interactions. (abstract) Bull. Amer. Phys. Soc. 61 (20): <http://meetings.aps.org/Meeting/DFD16/Session/G17.6>.
11. Paes, P.L.R., Xuan, Y., Brasseur, J.G. 2015 Numerical investigation of kinetic energy dynamics during autoignition of n-heptane/air mixture. (abstract) Bull. Amer. Phys. Soc. 60 (21): <http://meetings.aps.org/link/BAPS.2015.DFD.G40.6>.

4.4. Presentations

1. March 2020, Brasseur: Aerospace Engineering Sciences, University of Colorado: "Interscale Turbulence-Chemistry Dynamics with Reduced Basis Representations for Application to LES Modeling"
2. 73rd Annual Meeting of the APS Division of Fluid Dynamics, 23-26 November, 2020, Chicago/virtual: 1 presentation (see section 4.3, item 1)
3. 72nd Annual Meeting of the APS Division of Fluid Dynamics, 22-24 November, Seattle, WA, 2019: 1 presentation (see section 4.3, item 2)
4. 11th U.S. National Combustion Meeting, Pasadena, California, March 24-27, 2019: 1 presentation (see section 4.3, item 3)
5. AIAA SciTech Forum and Exposition 2019, San Diego, CA, 7-11 January 2019: 3 presentations (see section 4.1, items 5-7).
6. 71st Annual Meeting of the American Physical Society's Division of Fluid Dynamics, Atlanta, Georgia 18-20 November 2018: 3 presentations (see section 4.3, items 4-6).
7. 70th Annual Meeting of the APS Division of Fluid Dynamics, November 17-21, Denver, CO, November 2017: 2 presentations (see section 4.3, items 7-8)
8. 10th U. S. National Combustion Meeting, April 23-26, 2017, College Park, MD, 2017: 1 paper (see section 4.3, item 9)
9. AFOSR 2019 AFOSR/ARO/NSF Basic Combustion Research Review, 3-7 June 2019: review presentation.
10. AFOSR Computational Mathematics Research Review, 12-16 August 2019: review presentation.
11. AFOSR Computational Mathematics Research Review, 13-15 August 2018: review presentation plus presentation at the preceding Workshop on Challenges in Reduced Order Modeling of Combustion: "Issues on the Incorporation of Essential Physics in Closures of Nonlinearities in LES of Premixed Turbulent Combustion".

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- [1] Paes, P.L.K., Brasseur, J.G., Xuan, Y., Shah, Y.G. 2021 Fourier-Physical Space Coherent Structure in Flame-Vortex Interactions Relevant to Flame-turbulence Interactions using a New Signal Periodization Procedure, In press, *AIP Advances*, <https://doi.org/10.1063/5.0050280>.
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- [3] Paes, P.L.K., Shah, Y.G., Xuan, Y., Brasseur, J.G. 2019 Dynamically Dominant Interscale Couplings in the Nonlinear Chemical Source Terms for Species Evolution in Premixed Turbulent Combustion with Application to LES Modeling. 11th U.S. National Combustion Meeting, Pasadena, California, March 24-27, 2019.
- [4] Shah, Y.G., Brasseur, J.G., Xuan, Y. 2020 Assessment of disparities in estimating filtered chemical reaction rates in LES using DNS of turbulent premixed combustion. *Combustion Theory and Modelling*, <https://doi.org/10.1080/13647830.2020.1831616>.
- [5] Shah, Y.G., Brasseur, J.G., Xuan, Y., 2021 On the Structure of Local Subfilter-scale Species Surrounding Flame Fronts in LES of Premixed Turbulent Combustion, *AIP Advances*, in press, *Physics of Fluids*, <https://doi.org/10.1063/5.0045870>.
- [6] Paes, P.L.K., Xuan, Y., Shah, Y.G., Brasseur, J.G. 2019c Removing Pollution in the Fourier Space Representation of Non-Periodic Signals over Finite Domains, AIAA Paper No. 2019-0903, January 2019 SciTech Meeting, San Diego.
- [7] Paes, P.L.K., Brasseur, J.G., Xuan, Y. 2019b Kinematic Relationships between Physical and Fourier Space in Premixed Turbulent Combustion for Application to Large-Eddy Simulation. AIAA Paper No. 2019-2145, January 2019 SciTech Meeting, San Diego.

Appendix

Fourier-Physical Space Coherent Structure in Flame-Vortex Interactions Relevant to Flame-Turbulence Interactions using a New Signal Periodization Procedure

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(Dated: 17 March 2021)

The aim of the current study is to characterize key multidimensional relationships between coherent structure in physical vs. Fourier/scale space representations of flame-turbulence interactions, as a basis for future analysis of the nonlinear couplings between key resolved-scale (RS) and subfilter-scale (SFS) motions in large-eddy simulation (LES) of premixed turbulent combustion. However, to apply the bounded Fourier transform (FTF) in the nonperiodic flame-normal direction requires the removal of nonphysical Fourier content from the boundary discontinuities. To this end, we have developed a broadly applicable “discontinuity pollution removal” procedure for application to the FTF of multidimensional signals with a single nonperiodic direction. The procedure balances periodization of the signal near the boundaries with minimization of signal modification away from the boundaries. We applied the procedure in a physical-Fourier space analysis of the interactions between a flame and single-scale eddies modeled as the impact of a train of two-dimensional (2D) vortices on an initially planar premixed flame. We find that a specific spectrally-broad localized coherent structure in Fourier space connects RS to SFS fluctuations in thermal energy and species concentration that, in physical space, are localized to the corrugations in the flame front in response to eddy-flame interactions. Within the RS fluctuations of energy and species concentration, flame corrugation structure in physical space is found to be localized to sub volumes within the RS region of 2D Fourier space. This new understanding of physical-Fourier space relationships categorizes classes of RS-SFS interactions relevant to SFS modeling in LES of premixed turbulent combustion.

I. INTRODUCTION AND AIMS

The large-eddy simulation (LES) framework centers on decomposition in scale; LES modeling centers on the interactions between larger “resolved-scale” (RS) and smaller “subfilter-scale” (SFS) fluctuations in all dependent variables. When applied to premixed turbulent combustion, the reality of (practical) LES is that there necessarily exists key SFS chemical kinetics and thermo-mechanical dynamics that impact RS evolution but cannot be supported by the grid. Thus, unlike LES of incompressible turbulence, where only momentum dynamics are involved, LES modeling of strongly density-varying turbulent flows in the presence of chemical reaction and heat release must characterize key impacts of SFS fluctuations on the evolution of RS momentum, thermal energy, and concentrations of many chemically-reacting species. In particular, SFS modeling is required for advective and chemical nonlinearities that couple the unresolvable evolutions of SFS variables with the evolution of RS variables supported by the effective grid. (“Effective” implies the existence of additional coarsening of the geometric grid from implicit filtering due to dissipative content in the SFS models and numerical algorithm.) The central aim of our current research program is to ascertain the relationships between coherent structure in physical space and coherent structure in Fourier space with data from reduced physics simulations that are designed to

incorporate key dynamics from flame-turbulence interactions, ubiquitous in premixed turbulent combustion. As discussed in more detail in Sec. III A, this knowledge will be later used in analyses of key nonlinear RS-SFS couplings central to the evolution of coherent features of RS momentum, energy and species concentration.

Our analysis centers on scale-based decomposition of the key dynamic variables. Decompositions such as proper orthogonal decomposition (POD)^{1,2} and dynamic mode decomposition (DMD)³ are therefore not useful for our needs. We choose the multidimensional Fourier decomposition for two primary reasons. Firstly, the Fourier transform (FTF) is useful to decompose multidimensional multiscale analytic functions into scale-based series of complex exponentials that are mathematically convenient for scale-based analysis in elemental form. The wavelet transform, for example, is a filtered version of the Fourier decomposition with Fourier modes grouped according to the choice of wavelet filter⁴. In multidimensional Fourier space, a Fourier mode is a complex harmonic function that is quantified with precisely-defined scale (wavelength), direction (wavevector), amplitude and phase (Fourier coefficient). Fourier decomposition of a continuous multiscale signal is “elemental” in the sense that, whereas a single Fourier mode does not represent physical-space structure, collections of phase-correlated Fourier modes within localized volumes in Fourier space characterize scale-based coherent structure in physical space, for example through wavelet decomposition^{4–6}. Each Fourier mode in the collection provides an elemental contribution to scale, strength, direction, and structure, with coherence determined by the phase rela-

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tionships among the collection of Fourier modes⁴.

Multiscale nonlinear dynamical systems such as turbulence generally describe characteristic “scales of motion” statistically⁷. Local space-time analysis is required to relate spatial scales to coherent structural elements such as vortex tubes and sheets, and local concentrations of turbulent kinetic energy, strain-rate, and vorticity⁸. However, once the local relationships of scale-based turbulence structure between Fourier and physical space are understood, Fourier-based scale decomposition becomes a valuable tool to analyze scale-based coherent structure and nonlinear interscale interactions in scale space in relationship to nonlinear interactions between scale-dependent coherent structure in physical space⁹.

Future publications will apply knowledge from the current, kinematic, study to analyze key dynamical nonlinearities in the equations of motion¹⁰. This direction for analysis creates the second, more important, reason for our application of Fourier scale decomposition: representation of a nonlinearity underlying interscale dynamics at its most elemental level. The FTF of the advective nonlinearities in the governing equations for momentum, energy, and species concentration in turbulent combustion, for example, appear as linear sums of elemental nonlinear “triadic” interactions among three Fourier modes with wave-vectors that form a closed triangle in multidimensional Fourier space^{9,11,12}. In related studies^{10,13,14}, we make use of this Fourier-spectral property to analyze the nonlinear interactions among scales of motion within turbulent-reacting flows in context with LES, where turbulence structure is separated into RS and SFS spectral content. In the current study, we focus on the kinematic Fourier-physical-space relationships that underlie RS and SFS structure in flame-turbulence interactions.

Alternative approaches to scale-based decomposition apply basis functions that are mathematically more complex than the elemental Fourier basis. Examples include wavelet^{15,16} and empirical mode¹⁷ decomposition. The mathematical complexity of these basis functions, however, produces complicated representations of nonlinear terms in the transformed equations that greatly increase analytical complexity and reduce generality. The mathematical representation of nonlinear interactions among wavelet modes, for example, is a great deal more complex than the representations of elemental interactions among individual Fourier modes^{18,19}. However, the wavelet transform of nonlinearities can be equivalently represented in Fourier space as sums of elemental nonlinear terms among filtered groupings of Fourier modes in localized volumes within multidimensional Fourier space. In that way, one can quantify the dynamical couplings between regions kinematically associated with different coherent features of the dynamical system. This is the approach we take here.

However, to develop this analysis one must recognize and eliminate spurious spectral content that is introduced when the FTF is applied to a signal that is nonperiodic over a bounded domain. This is because the FTF of a bounded nonperiodic continuous analytic signal produces periodic extensions of the signal in physical space with discontinuities at the boundaries. As a global basis, the corresponding Fourier coefficients in-

clude contributions from these boundary discontinuities. In practice, signals from experimental measurements and numerical simulations are generally nonperiodic in at least one coordinate direction. Numerical simulations of flame-turbulence interactions in premixed turbulence combustion, for example, typically apply periodic boundary conditions in directions transverse to the mean flow, representing turbulence as statistically homogeneous in those directions; boundary discontinuities therefore do not exist in those directions. In contrast, the flame-normal direction the turbulence is statistically inhomogeneous and nonperiodic inflow/outflow boundary conditions are typically applied, creating boundary discontinuities in the periodically extended signal. However, computational and experimental signals are discretized, requiring the application of the discrete Fourier transform (DFT). The boundary “discontinuity” then becomes a large gradient with magnitude given by the jump in the signal between the inflow and outflow boundaries divided by the discretization scale. We shall continue to use the term “boundary discontinuity” to refer to the boundary gradients in discretized signals that characterize mathematical discontinuities in the corresponding continuous signal.

Nonperiodicity introduces an undesirable artifact in the Fourier transform that complicates scale-based analysis: each boundary discontinuity creates Fourier coefficients with power law decay extending from lowest to highest wavenumber in the direction of the discontinuity. Consequently, when the highest physical gradients are resolved by the grid, the power-law artifact from the boundary discontinuity dominates the energy content of the physical signal at high wavenumbers. In the image processing literature²⁰, the FTF of two-dimensional (2D) signals creates two power-law signatures referred to as “cross structure.” Several methods have been proposed to overcome or minimize the impacts of this unwanted spectral content (“pollution”) from boundary discontinuities that overlaps the signal of interest in Fourier space (see Sec. II A). These methods are typically described as “periodization” procedures.

Classical methods to periodize signals include the application of smooth physical-space filters at the non periodic boundaries²¹ and the antisymmetric signal extension²² of the signal. Foucaut *et al.*²³ developed a method to remove the boundary discontinuities of experimental Particle Image Velocimetry (PIV) data by removing a straight line connecting first and last sample points of the signal and imposing antisymmetry around the last sample point. Bruno *et al.*²⁴ developed a “Fourier continuation” method to periodize a signal within the Fourier framework by creating Fourier series within domains larger than the domain of the signal. Moisan²⁵ proposed a signal decomposition into a periodic component that resembles the original signal and a smooth component that captures the boundary discontinuities with linear functions within the domain, a step related to Foucaut *et al.*²³ as well as to the approach presented in Sec. II.

Whereas the methods described above retain spectral accuracy while removing spurious scale content by fully periodizing the signal, they also introduce new nonphysical Fourier content that overlaps with that of the original signal, making

these approaches unsuitable for our applications. In particular, smooth filters at the non periodic boundaries introduce new spectral content associated with widening filter. As we shall show (Sec. II B and II C), while the Foucaut *et al.*²³ and Moisan²⁵ approaches fully periodize the signal, they also modify the signal within the domain of interest at levels that are unacceptable for our application. Furthermore, whereas the antisymmetric signal extension removes C^0 discontinuities at the boundaries, it also introduces C^1 boundary discontinuities and makes the Fourier coefficients real-valued, masking phase information contained in the Fourier coefficients of the original signal and creating overlaps that cannot be removed. The “Fourier continuation” method introduces nonphysical large-scale Fourier content associated with the extension of the signal beyond its original domain, which overlaps with scales of interest in the original signal.

To overcome inadequacies of existing periodization methods for our application, we have developed a “pollution removal” methodology that specifically treats the tradeoff between full periodization and alteration of the signal in regions of interest (Sec. II). We present a semi-empirical approach that removes pollution from boundary discontinuities without creating additional spurious content that overlaps the signal in regions of interest in both Fourier and physical space. This is done through user-based empiricism introduced to balance between the need to modify the signal near the boundaries for periodization with the extent to which the signal is modified away from the boundaries. We extend our method to higher order discontinuities in Appendix A, and to multidimensional signals in Appendix B.

The current study aims to identify key Fourier-physical space relationships between coherent features in canonical flame-turbulence interactions. As discussed in Sec. III A, after applying the discontinuity pollution approach described here, we analyze the relationships between coherent structure in physical vs. Fourier space using a reduced physics simulation that is designed to characterize key dynamical interactions between a chemically-reacting flame and trains of single-scale eddies represented as 2D vortices. The specific objectives of the current paper are (1) to identify the spurious Fourier content created from nonphysical boundary discontinuities when the FTF is taken of nonperiodic variables on a bounded domain and develop a procedure to systematically remove the spurious content (“pollution”) from the boundary discontinuities without significantly altering the signal away from the boundaries, (2) to develop a reduced physics simulation of the interaction between an initially unstretched flame and a train of vortices and apply the simulation to validate the discontinuity-pollution-removal method, and (3) to analyze the 2D flame-vortex interaction model concurrently in Fourier and physical space to characterize the kinematic relationships between localized coherent structure in physical space and localized coherent structure in Fourier space within the LES framework.

The paper is organized as follows: Section II presents the mathematical representation underlying the Fourier description of nonperiodic signals over finite domains and the proposed “discontinuity pollution removal” procedure for one-

dimensional (1D) signals. The procedure is extended to multidimensional signals in Appendix B and in Appendix C validated using reduced-physics simulations of 2D flame-vortex-train interactions (refereed henceforth as flame-vortex interactions). In Sec. III A we present the concepts underlying the applicability of the flame-vortex interaction model to flame-turbulence interactions before describing the reduced physics model in Sec. III B. This is followed in Sec. III C with an analysis of coherent structure of flame-vortex interactions in physical space. The Fourier space counterpart of coherent structure analysis is presented in Sec. IV using systematic spectral filtering. The relative contributions of distinct physical-Fourier space coherent structures to the energy content of distinct signals in context with the LES framework are analyzed in Sec. IV D. Key findings are discussed in Sec. V.

II. REMOVAL OF POLLUTION IN THE FOURIER TRANSFORM OF NONPERIODIC SIGNALS OVER FINITE DOMAINS

In this section, we make use of Fourier theory to provide a better understanding of issues and implications of Fourier decomposition of nonperiodic signals over a finite domain. This analysis provides the foundation for the proposed “discontinuity pollution removal” procedure.

A. The Issues with Fourier Decomposition of Nonperiodic Signals

As discussed in Sec. I, because the Fourier basis is unbounded, the FTF of a continuous function on a bounded domain includes periodic extensions of the function to infinity in all coordinate directions. In directions where the signal is not periodic, therefore, the periodically extended signal includes discontinuities at the boundaries that are reflected in the complete set of discrete Fourier coefficients. When the discrete FTF is applied to discretized nonperiodic functions, the infinite derivatives at the boundary discontinuities are now large-but-finite derivatives that are determined by the extent of jump together with grid resolution at the boundaries. The discontinuities at the boundaries between the function and its periodic extension may appear at the function level (C^0), but also in higher order derivatives $f^{(m)}(x) = d^m f/dx^m$ with $m \geq 1$, where superscript (m) indicates the derivative.

To illustrate, consider the 1D bounded function $f(x)$ shown in Fig. 1, constructed by combining a cosine function with characteristic length scale ℓ with a Gaussian function with length scale η . With this function we create three 1D signals defined over finite domains $L_1 = 6\ell$, $L_2 = 4.998\ell$, and $L_3 = 5.598\ell$, which are indicated as f_1 , f_2 , and f_3 , respectively. $\eta = L_1/36$, and $\xi = L_1/3$ for all three signals. Figures 1a-c show periodic extensions of f_1 , f_2 , and f_3 , where the solid black vertical lines indicate the periodically extended domain boundaries. By construction, the three signals have the same characteristic length scales but different finite domain sizes.

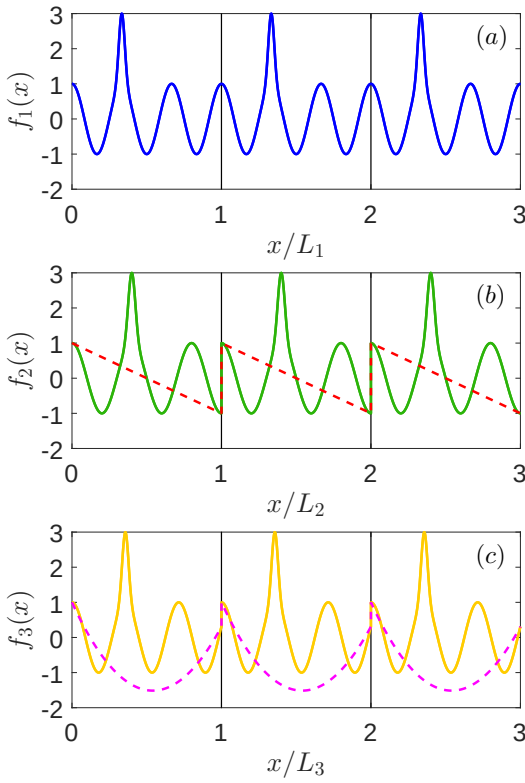


FIG. 1: Periodic extension of (a) f_1 , (b) f_2 , and (c) f_3 , where $f(x) = \cos(\pi x/\ell) + 2e^{-(x-\xi)/\eta}$, $L_1 = 6\ell$, $L_2 = 4.998\ell$, $L_3 = 5.598\ell$, $\eta = \ell/6 = L_1/36$, and $\xi = 2\ell = L_1/3$. The dashed red lines in (b) indicate the C^0 discontinuity function given by Eq. (3). The dashed magenta curve in (c) indicate the C^0 and C^1 boundary discontinuities derived in Appendix A.

The three signals in Fig. 1 are designed to characterize three different levels of discontinuity: (a) is a periodic signal that is continuous in the function at all its derivatives at the boundaries, (b) is a nonperiodic signal with only a C^0 discontinuity at the boundaries, and (c) is a nonperiodic signal with C^0 and C^1 boundary discontinuities. The jumps in boundary values (“discontinuities”) in the periodic extension of nonperiodic signals f_2 and f_3 are indicated by vertical dashed lines at the boundaries in Figs. 1b and c, respectively. These represent the boundary discontinuities at the function level C^0 . f_3 discontinuities has both C^0 and C^1 with jumps in boundary values of f and its derivative $f^{(1)}(x) = df/dx$.

The Fourier space spectrum of f_1 , f_2 , and f_3 in Fig. 1a-c is shown in Fig. 2 where the wavenumber axis (k_x) is nondimensionalized with the cosine length scale ℓ . The Fourier transform of the periodic signal (the blue curve in Fig. 2) has a spectral peak at $k_x\ell/\pi = 1$ and an exponential drop above the scale η of the Gaussian function ($k_x\ell/\pi = \ell/\eta = 6$). In contrast, the Fourier content of signals f_2 and f_3 with nonperiodic boundary values (green and yellow curves in Fig. 2, respectively) have clear power-law decay of squared Fourier coefficients towards high wavenumbers with non-monotonic behavior surrounding a broad peak around $k_x\ell/\pi = 1$. We shall find

that the difference between $\hat{f}_2^2$ and $\hat{f}_3^2$ at low wavenumbers ($k_x\ell/\pi \lesssim 4$) reflects the existence of the C^1 boundary discontinuity in f_3 .

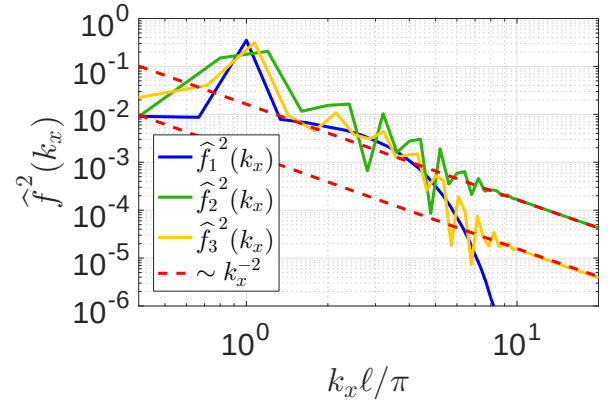


FIG. 2: Squared Fourier coefficients $\hat{f}_1^2$ and $\hat{f}_2^2$ over wavenumber axis k_x nondimensionalized by the scale ℓ of the cosine function (see Fig. 1). The dashed red line indicates the C^0 boundary discontinuity given by Eq. (2).

As shown in what follows, the boundary discontinuities presented in Fig. 1b-c have broad-band power-law Fourier energy content that overwhelms and pollutes the Fourier content of the original signal at scales of interest. In Fig. 2, for example, the power-law contribution from the C^0 boundary discontinuity in f_2 completely obscures the true signal f_1 at scales below the scale of the Gaussian function (i.e., $k_x\ell/\pi \gtrsim 6$) and pollutes the true signal at lower wavenumbers. Note that the discretization of the signal required to apply the FFT was chosen so that the cutoff in the power-law behavior at the discretization scale occurred at k_x outside the range plotted in Fig. 2. Our aim is to remove the boundary-generated pollution shown in Fig. 2 without significantly interfering with the underlying signal of interest (blue curve) that we aim to recover in the pollution removal process.

To advance this aim, we quantify the Fourier coefficients of the boundary discontinuity from a nonperiodic signal. Consider an arbitrary smooth continuous 1D signal $f(x)$ over a finite domain $0 \leq x \leq L$ with boundary values $f(0)$ and $f(L)$. The periodic extension of $f(x)$ has a C^0 boundary discontinuity with boundary jump $\Delta f = f(L) - f(0)$. The bounded FTF of $f(x)$ is

$$\hat{f}(k_x \neq 0) = \frac{1}{L} \int_0^L f(x) e^{-ik_x x} dx = -\frac{i}{k_x} \hat{f}'(k_x) + \frac{i}{k_x} \frac{\Delta f}{L}, \quad (1)$$

where $\hat{f}(k_x)$ are the Fourier coefficients of $f(x)$ and $\hat{f}'(k_x)$ are the Fourier coefficients of $f' = df/dx$, each a function of the infinite set of discrete wavenumbers, $k_x = k_n = 2\pi n/L$, $n = \pm 1, \dots, \pm \infty$. $k_x = 0$ is the average of $f(x)$ over the finite domain. The RHS of Eq. (1) was obtained using integration by parts and $e^{-ik_x L} = e^{-i2\pi n} = 1$.

The second term on the RHS of Eq. (1) isolates the Fourier content associated with the C^0 discontinuity in the periodic extension of $f(x)$. For periodic signals ($\Delta f = 0$), the second

term does not contribute to the derivative of $f(x)$. For nonperiodic signals, the magnitude of this term follows a k_x^{-1} power law with amplitude proportional to Δf . Thus, the fundamental C^0 discontinuity function in Fourier space is described by

$$\widehat{d}(k_x) = \frac{i}{k_x} \frac{\Delta f}{L}, \text{ for } k_x \neq 0. \quad (2)$$

$\widehat{d}(0) = 0$ so that the discontinuity function does not contribute to the mean. The inverse FTF of $\widehat{d}(k_x)$ within the finite domain is

$$d(x) = \frac{\Delta f}{L} \left(x - \frac{L}{2} \right), \quad (3)$$

where $0 \leq x \leq L$. The physical space counterpart of Eq. (2) captures the fundamental C^0 discontinuity in $f(x)$ at the domain boundaries ($\Delta d = \Delta f$) with a linear sawtooth function extended periodically to infinity. The discontinuity function in physical and Fourier space is indicated by dashed red lines in Fig. 1b and Fig. 2 (with the mean included in $d(x)$ for better comparison with $f(x)$).

The linear sawtooth function in Eq. (3) captures only the C^0 boundary discontinuity in $f(x)$. In Appendix A, Eqs. 1-3 are generalized to C^n discontinuities in the higher order derivatives $f^{(n)}(x) = d^n f/dx^n$ with $n = 0, 1, \dots, N$. Like the discontinuity function for $f(x)$ given by Eq. (1), the discontinuity in the highest order derivative $f^{(N)}$ is represented in physical space by a linear discontinuity function $d_N^{(N)}(x)$ given by Eq. (A1). This function is sequentially integrated back to the 0^{th} derivative level ($n = 0$) to produce an N^{th} -order polynomial $d_N^{(0)}(x)$ that includes all discontinuities in $f^{(n)}(x)$ from the function level $n = 0$ up to the derivative level $n = N$. For example, a boundary discontinuity in $f^{(1)}(x) = df/dx$ is described by the linear discontinuity function $d_1^{(1)}(x)$ given by Eq. (A4) that when integrated to the function level produces the discontinuity function $d_1^{(0)}(x)$ given by Eq. (A6), the sum of parabolic plus linear functions.

In Fourier space, the sequential integration of $d_N^{(N)}(x)$ back to the $n = 0$ derivative level produces Fourier coefficients for $d_N^{(0)}(x)$ that follow a sum of power laws $\sim k_x^{-(n+1)}$ with $n = 0, 1, \dots, N$, given by Eq. (A12). The amplitude of each power law is proportional to the magnitude of the boundary jump $\Delta f^{(n)} \equiv f^{(n)}(L) - f^{(n)}(0)$ at each derivative level, n . For example, the Fourier description of a function $f(x)$ with boundary discontinuities up to the 1^{st} derivative level is given by (see Eq. (A12)):

$$\widehat{d}_1^{(0)}(k_x \neq 0) = \frac{\Delta f^{(0)}}{L} \left(\frac{i}{k_x} \right) + \frac{\Delta f^{(1)}}{L} \left(\frac{i}{k_x} \right)^2, \quad (4)$$

where $\Delta f^{(0)} = \Delta f$.

As an example, Fig. 3 shows the physical and Fourier space views of the C^0 and C^1 discontinuity functions of f_3 shown in Figs. 1c and 2. The boundary discontinuities are represented by mathematical expressions for $d_0^{(0)}(x)$ and $d_1^{(0)}(x)$ from boundary discontinuities in both the function $f(x)$ and

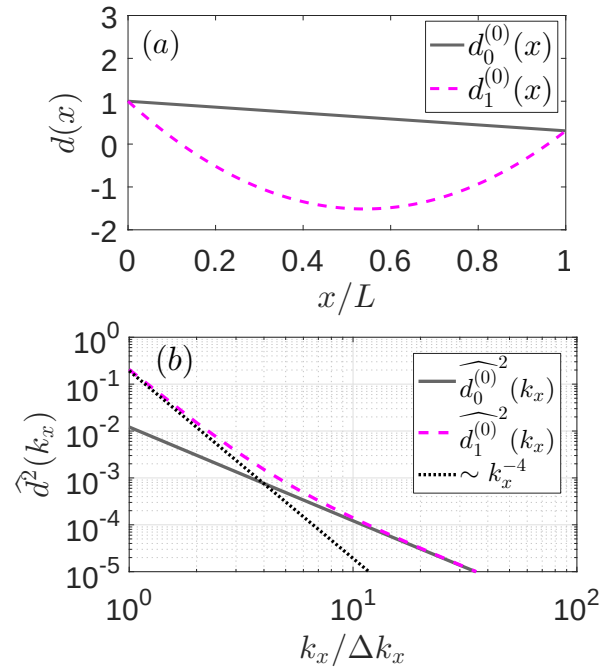


FIG. 3: (a) The physical space function and (b) Fourier space spectral content of $d_0^{(0)}(x)$ and $d_1^{(0)}(x)$ derived in Appendix A to capture the C^0 and $C^0 + C^1$ boundary discontinuities for $f_3(x)$ shown in Fig. 1c having $\Delta f^{(0)} = -0.69$, $\Delta f^{(1)} L_3 = 17.23$.

its first derivative $f^{(1)}(x)$ (see Appendix A), that is for both C^0 (Eqs. 2 and 3) and $C^0 + C^1$ (Eqs. 4 and A6) boundary discontinuities in $f(x)$. In Fourier space (Fig. 3b), the squared Fourier coefficients of $d_1^{(0)}(x)$ follow a power-law $\sim k_x^{-2}$ in the high wavenumber limit and a different power law $\sim k_x^{-4}$ in the low wavenumber limit, each associated with the Fourier representations of C^0 and C^1 discontinuity functions, respectively. In physical space (Fig. 3a), the parabolic function $d_1^{(0)}(x)$ captures the fundamental boundary discontinuities C^0 and C^1 . Higher order boundary discontinuities do not exist since the function is smooth in derivatives above one. The linear function $d_0^{(0)}(x)$ captures only the fundamental boundary discontinuity C^0 . The polynomials $d_N^{(0)}$ are constructed to capture the fundamental boundary discontinuity only up to C^N .

The fact that the higher order boundary discontinuities contain higher power-law exponents in $1/k_x$ will turn out to be important in the application of the discontinuity pollution removal procedure discussed in the next sections, where the discontinuity removal function $d_N^{(0)}(x)$ will be used to remove pollution in Fourier space associated with the boundary discontinuities in the function. As shown in Fig. 3b, for example, the variance of the discontinuity removal function for a function with discontinuity in the function and its first derivative is dominated by the C^1 discontinuity at wavenumbers below 4 and the C^0 discontinuity at high wavenumbers. We shall show that discontinuity removal should be limited to higher wavenumbers where the $d_0^{(0)}$ discontinuity dominates by or-

ders of magnitude over higher-order singularities. Thus, in practice the pollution removal procedure generally needs only be applied to the C^0 boundary discontinuity. That is the case in the current study.

B. Pollution in the Fourier Representation of a Physical Nonperiodic Signal

In Fig. 4 we illustrate spectral pollution with a physically relevant 1D signal obtained from a 2D simulation described in Sec. III B of the interaction between a laminar flame and a train of counter-rotating 2D vortices. Inflow/outflow boundary conditions are applied across a domain length of $16D$, where D is the diameter of the individual vortices within the train. The blue curve in Fig. 4a is the streamwise momentum component along a pencil between the inflow and outflow boundary. The flame, identified by the peak at $x/D = 4$, produces the largest physical gradient at the smallest physical scale, the flame scale, δ_f . In Fig. 4b we plot, with the blue curve, the Fourier spectral content of the bounded 1D signal in Fig. 4a (blue curve).

As discussed above, Fig. 4 demonstrates that the bounded FTF of a continuous nonperiodic signal produces C^0 boundary discontinuities in physical space with corresponding k^{-2} broadband spectral energy content from the first wavenumber to $k_{max} \approx \pi/\Delta x$ (Eq. (2)). Direct Numerical Simulation (DNS) of turbulence requires that Δx resolves well the smallest dynamically-relevant viscous scale, so that the physical signal has a viscous spectral roll-off that initiates at a scale an order of magnitude larger than the grid scale. As shown by the blue curve in Fig. 4b, this roll-off is masked by the power-law content of the C^0 boundary discontinuity that overwhelms the frictional scale content at higher wavenumbers. The aim is to remove this pollution and uncover the small-scale content masked by the C^0 power law at high wavenumbers.

If one simply removed $\hat{d}(k_x)$ given by Eq. (2) from the discrete FTF of the original nonperiodic signal $f(x)$, the inverse transform $\hat{f}(k_x) - \hat{d}(k_x)$ would produce a periodized form of the blue signal in Fig. 4 with the fundamental C^0 boundary discontinuity removed. This fully periodized signal is shown in physical space by the solid green curve in Fig. 4a, the original blue curve with the sawtooth function in Fig. 4a subtracted (after shifting to zero mean). Because the sawtooth function includes the C^0 boundary discontinuity, the full removal of pollution from the boundary discontinuity is equivalent to full periodization, as noted in^{23,25}.

The squared Fourier coefficients of the periodized signal are shown in Fig. 4b with the solid green curve. Note that the curve has an exponential drop off beginning near the wavenumber corresponding to the flame length scale, indicating that the pollution at high wavenumbers corresponding to the discontinuity has been removed. This is particularly obvious at wavenumbers $k_x \delta_f / \pi \gtrsim 1$ where the k^{-2} reduction in $\hat{f}^2(k_x)$ dominates and fully masks the true signal. At wavenumbers

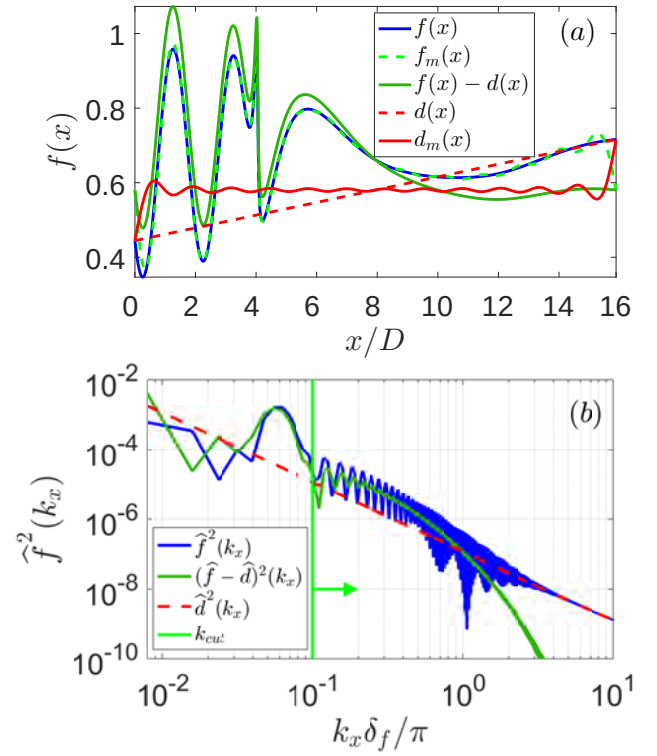


FIG. 4: (a) Physical and (b) Fourier space view of 1D signal extracted from 2D flame-vortex simulations described in III B. Blue curves show the original signal, dashed red curves indicate the expressions that capture the fundamental C^0 boundary discontinuities (Eqs. (3) and (2)), and dark green curves show the “periodized” signal after removing the later from the former. The mean $f(x)$ is added to $d(x)$ for better comparison. The dashed green curve in (a) indicate post-processed signal discussed in Sec. II C using the filter cutoff wavenumber k_{cut} indicated by vertical green line in (b).

below $k_x \delta_f / \pi \approx 1$ the Fourier coefficients associated with the signal dominate the coefficients associated with the discontinuity so that the signal is modified relatively little there. However, at wavenumbers $k_x \delta_f / \pi \lesssim 0.05$, the lowest four wavenumbers, the Fourier coefficients within the power-law drop contain variance that overlaps with the true signal. This modification at the lowest few wavenumbers result from subtracting the “ramp” part of the sawtooth function, the largest scale feature of the inverse transform of the discontinuity term $\hat{d}(k_x)$ in Eq. (2). This content interferes with the true signal and modifies the overall shape of the function as shown by comparing the blue and dark green curves in Fig. 4a.

We find that in order to remove the C^0 boundary discontinuity from the signal $f(x)$ using the same C^0 discontinuity embedded in the sawtooth function $d(x)$, it was necessary to also change the signal globally over the domain. We further find from Fig. 4 that the extent of signal modification that is required to periodize the signal depends on the relative variance content of the signal vs. sawtooth function at the lowest wavenumbers, since the magnitude of the Fourier coefficients increases like k_x^{-2} . The lowest k_x content that defines the over-

all shape of the largest-scale variations in the signal. In effect, by removing pollution at the highest wavenumbers that results from the C^0 boundary discontinuity from non-periodicity in $f(x)$, pollution is added at the lowest wavenumbers to account for the signal content in $d(x)$ between the boundaries, at the very largest Fourier scales. In the next section, we introduce a procedure for removing spurious signal content at the highest wavenumbers arising from the boundary discontinuity within $d(x)$ while minimizing modification of the signal of interest at the lowest wavenumbers. To do so requires a tradeoff that must be handled empirically.

C. A Proposed Pollution Removal Procedure that balances Periodicity with Signal Modification

Since the boundary discontinuity is associated with the highest wavenumbers, consider a high-pass-filtered version of the discontinuity function:

$$\hat{d}_m(k_x; k_{cut}) = m(k_x; k_{cut}) \hat{d}(k_x), \quad (5)$$

where

$$m(k_x; k_{cut}) = \begin{cases} 1 & \text{if } |k_x| \geq k_{cut} \\ 0 & \text{if } |k_x| < k_{cut} \end{cases}. \quad (6)$$

The inverse transform of $\hat{d}_m(k_x; k_{cut})$, $d_m(x; k_{cut})$, is given in Fig. 5a for the three filter scales k_{cut} shown in Fig. 5b. The red dashed curve in Fig. 5a includes all Fourier coefficients ($k_{cut} = 0$). As k_{cut} is systematically moved towards higher wavenumbers, $d_m(x; k_{cut})$ systematically localizes around the discontinuity at the domain boundaries the contribution from the center of the domain.

The concept is to modulate $f(x)$ by subtracting $d_m(x; k_{cut})$ rather than $d(x)$ to create the modulated function $f_m(x; k_{cut}) = f(x) - d_m(x; k_{cut})$. As just discussed, when $k_{cut} = 0$ all Fourier modes of $f(x)$ are modulated ($d_m(x; k_{cut}) = d(x)$), the boundary discontinuity is fully removed and the signal is fully periodized; however, the modulated version $f(x) - d(x)$ has been modulated globally. In contrast, when $k_{cut} = k_{max}$, $f(x)$ is left unchanged ($d_m(x; k_{cut}) = 0$) and the signal remains fully non-periodic. It is therefore not surprising to find that the jump in the modulated signal $f_m(x; k_{cut})$ depends on k_{cut} . This is shown in the inset to Fig. 5a, where the degree of reduction in the boundary discontinuity $d_m(x; k_{cut})$ is shown to decrease with increasing k_{cut} . We note that the reduction is minimal when $k_{cut} \delta_f / \pi = 0.5$, just below the roll-off in the signal which, as shown in Fig. 4b, occurs at $k_x \delta_f / \pi \approx 1$. We conclude that there is a tradeoff between the degree of reduction in the boundary discontinuity and periodization of the signal (pushing k_{cut} to smaller values), and the minimization of signal modification away from the boundaries (pushing k_{cut} to higher values).

In order to quantify the interplay between the degree of periodization vs. the degree of modification of the original signal after removing $d_m(x; k_{cut})$ from $f(x)$, we define two parameters to quantify these two modulations. We then apply these

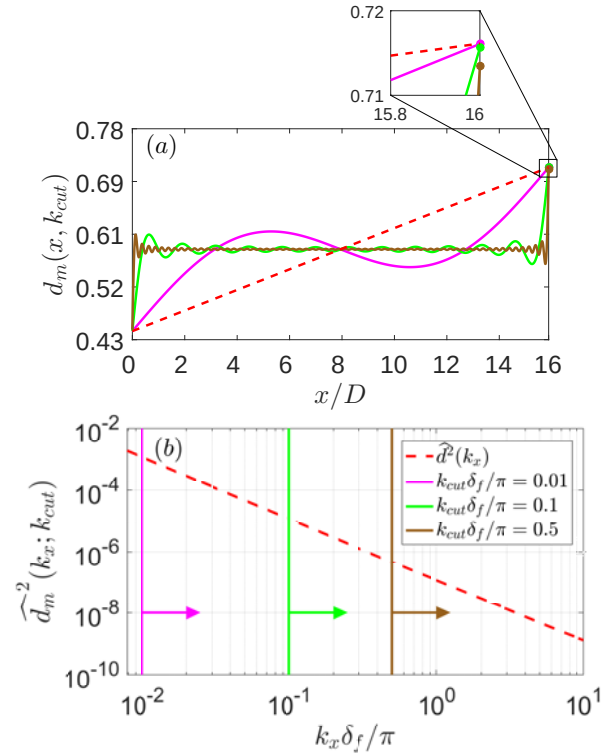


FIG. 5: (a) Physical and (b) Fourier space representation of modulated discontinuity $d_m(x; k_{cut})$ and $\hat{d}_m(k_x; k_{cut})$, respectively, for three cutoff wavenumbers $k_{cut} > 0$ within the power-law spectrum of $\hat{d}^2(k_x)$. The mean $f(x)$ is added to $d_m(x; k_{cut})$ for better comparison with Fig. 4. The inset of Fig. 5a shows boundary values for each $d_m(x; k_{cut})$.

two parameters within a “partial periodization” procedure that balances signal modulation with discontinuity removal. The first parameter is the ratio of content in the modulation function, $d_m(x; k_{cut})$ to the content in the original signal, $f(x)$, where “content” is defined with the L^2 norm:

$$\varepsilon(k_{cut}) = \frac{\|d_m(x; k_{cut})\|_V}{\|f(x)\|_V}. \quad (7)$$

The volume of interest, V , may be chosen as the entire domain or a subset of the domain, for example to exclude boundary regions. $\varepsilon(k_{cut})$ quantifies the relative variance content removed from the original signal. In principle, the aim is to minimize this parameter. The second parameter quantifies the level of residual gradient at the boundary relative to the maximum physical gradient within the volume of interest:

$$\Delta_r(k_{cut}) = \frac{|\Delta f_m(k_{cut})| / \Delta x}{\max_x (\partial f(x) / \partial x)_V}, \quad (8)$$

where

$$\Delta f_m(k_{cut}) = f_m(L, k_{cut}) - f_m(0, k_{cut}) = \Delta f - \Delta d_m(k_{cut}). \quad (9)$$

$\Delta f_m(k_{cut})$ is the residual boundary jump in the modified signal and Δf and $\Delta d_m(k_{cut})$ are the boundary jumps in the original

signal and modified discontinuity function, respectively. Like $\epsilon(k_{cut})$, the aim is to minimize $\Delta_r(k_{cut})$.

Consider Fig. 6, where the signal modification parameter $\epsilon(k_{cut})$ and the residual jump parameter $\Delta_r(k_{cut})$ are plotted against nondimensional k_{cut} for the physical signal shown in Fig. 4. The dashed vertical lines indicate the k_{cut} associated with each $d_m(x; k_{cut})$ shown in Fig. 5 and the symbols indicate their respective $\epsilon(k_{cut})$ and $\Delta_r(k_{cut})$ values. Both curves display approximate power-law dependencies that show the quantitative tradeoff between minimization of the boundary discontinuity (smaller k_{cut}) and minimization of signal modification (larger k_{cut}).

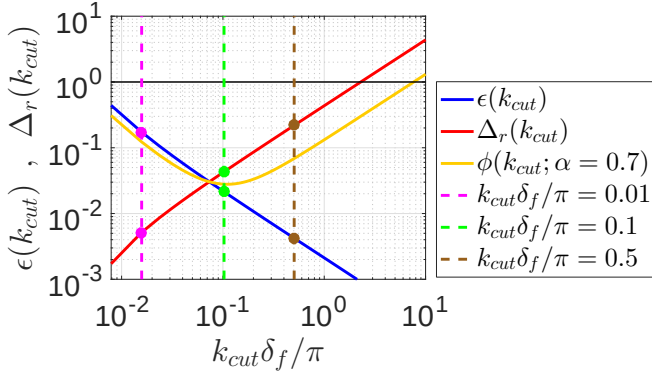


FIG. 6: Signal modification $\epsilon(k_{cut})$ and residual discontinuity $\Delta_r(k_{cut})$ as a function of cutoff wavenumber k_{cut} for the physical signal shown in Fig. 4.

The fact that $\Delta_r(k_{cut})$, the red curve in Fig. 6, exceeds 1 at the highest values of k_{cut} shows that the spurious gradient in $f(x)$ at the domain boundary exceeds the maximum physical gradient in $f(x)$ within domain, reflecting major pollution in Fourier space at high wavenumbers where in Fig. 4b the blue curve greatly exceeds the green curve. At these values of k_{cut} the modified signal retains strong non-periodicity. In contrast, at the lowest value of k_{cut} in Fig. 6, the red curve shows that the boundary discontinuity has been nearly entirely removed ($\Delta_r \approx 0.001$) and the modified signal is close to fully periodic, however the blue curve shows major modifications to the original signal ($\epsilon \approx 0.45$). However, at the crossover of the two curves, both the level of modification to the original function and the residual level of boundary discontinuity are reasonable, around 3%, suggesting that reasonable tradeoffs exist that can minimize both the pollution due to nonperiodicity and the degree of modification of the signal away from the boundaries, albeit with small residual levels of both remaining. This observation provides the basis for our proposed discontinuity pollution removal procedure.

The fundamental boundary discontinuity $\Delta f = f(L) - f(0)$ is represented in physical space by Eq. (3) and Fourier space by Eq. (2), a sawtooth function with zero mean and boundary discontinuities $\Delta d = d(L) - d(0)$ matching that of the function $f(x)$. The procedure to remove the discontinuity from the Fourier transform of $f(x)$ centers on the removal of Fourier content $\hat{d}_m(k_x; k_{cut}) = m(k_x; k_{cut})\hat{d}(k_x)$ from the original signal $\hat{f}(k_x)$, where $m(k_x; k_{cut})$ is a high-pass filter over

$k_x \geq k_{cut}$. Since the aim is to significantly modulate the signal only near the boundary discontinuity, $m(k_x; k_{cut})$ is designed to modify the signal over a to-be-determined range of high wavenumbers that localizes to the boundary discontinuity to create $f_m(x; k_{cut}) \leftrightarrow \hat{f}_m(k_x; k_{cut}) = \hat{f}(k_x) - m(k_x; k_{cut})\hat{d}(k_x)$.

The aim is to remove spectral content associated with the boundary discontinuity without significant modification of the signal away from the boundaries. To identify an optimal filter cutoff wavenumber k_{cut}^{opt} that provides the best tradeoff between minimization of signal modification, ϵ and minimization of the residual boundary discontinuity, Δ_r , we define the following weighted variable:

$$\phi(k_{cut}; \alpha) = \alpha \epsilon(k_{cut}) + (1 - \alpha) \Delta_r(k_{cut}), \quad (10)$$

where $0 < \alpha < 1$. α is a user-specific parameter that weights the relative contribution of ϵ vs. Δ_r . The “optimal” filter cutoff wavenumber k_{cut}^{opt} is defined as the minimum in $\phi(k_{cut}; \alpha)$ as a function of k_{cut} for specified fixed α :

$$\phi(k_{cut}^{opt}; \alpha) = \min_{k_{cut}} [\phi(k_{cut}; \alpha)]. \quad (11)$$

The choice of α depends on the characteristics of the original nonperiodic signal and the aims of the user. Large α favors minimization of changes in the signal, while small α the minimization of the residual boundary discontinuity and the retained level of periodicity. Note that ϵ and Δ_r (Eqs. 7 and 8) involve integrals over a user-specified volume, V , allowing the user to decide the region of the signal where modulation should be minimized. In this section, we use the entire domain for V .

To illustrate the tradeoffs in the selection of α , Fig. 7 shows the dependence of ϵ , Δ_r , and k_{cut}^{opt} for different choices of α using the 1D signal in Fig. 4. This figure shows that for each α there is a corresponding k_{cut}^{opt} from the minimization of $\phi(k_{cut}; \alpha)$, which is used to evaluate the tradeoffs between ϵ and Δ_r . For this signal if α were chosen as 0.7, for example (dashed line in Fig. 7), signal modification would be limited to 2% while the residual jump in the post-processed signal remains below 5%. The resulting weighted criteria curve $\phi(k_{cut}; \alpha = 0.7)$ is shown in Fig. 6 by the orange curve and the corresponding post-processed 1D signal using $k_{cut}^{opt} \delta_f / \pi = 0.1$ is indicated in Fig. 4a by the dashed green curve, $f_m(x)$.

In Appendix B the discontinuity pollution removal procedure is extended to three-dimensional (3D) signals with one nonperiodic direction and in Appendix C a sensitivity analysis of α is performed to exemplify the identification of α in practical applications.

III. COHERENT STRUCTURE IN FLAME-VORTEX INTERACTIONS AS A REDUCED PHYSICS MODEL OF FLAME-TURBULENCE INTERACTION

A. Underlying Concepts

Having developed a viable discontinuity pollution removal method, we are in a position to study scale-dependent coherent structure simultaneously in physical space and in Fourier

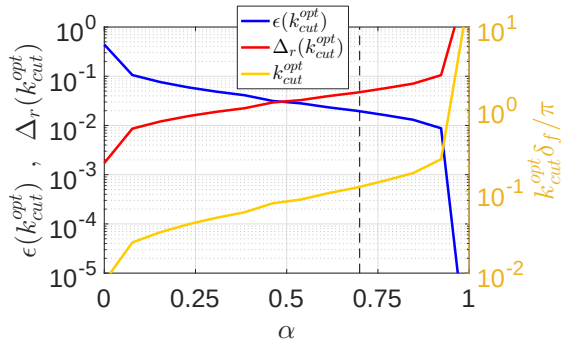


FIG. 7: ϵ (magenta), Δ_r (blue), and k_{cut}^{opt} (red) curves as a function of α for the 1D signal in Fig. 4 extracted from 2D flame-vortex interactions shown. The vertical dashed line is the value $\alpha = 0.7$ shown by the orange curve in Fig. 6.

scale space. A central aim of the current study is to determine the relationships among key coherent elements in momentum, thermal energy and species concentration from turbulence-flame interactions in terms of the characteristic scales of the coherent elements and their spatial inter-relationships concurrently in physical space and Fourier space. As discussed in Sec. I, we apply the Fourier spectral decomposition in part because the harmonic basis is explicitly scale-based, but specifically because the Fourier transform produces the most elemental description of the advective and chemical nonlinearities in the equations of motion as linear sums of nonlinear terms, each of which represents an elemental interaction among a few Fourier modes. As will be described in future manuscripts¹⁰, these mathematically elegant elemental forms within the Fourier spectral description of nonlinear interscale interactions may be rearranged in order to down-select a small percentage of the most dominant nonlinear interscale couplings that contribute to the evolution of resolved-scale momentum, thermal energy and species concentration predicted in LES of flame-turbulence interactions. These are the dominant dynamical interactions that underlie the modeled terms in LES of premixed turbulent combustion. The current study of the relationship between coherent structure in physical and Fourier space provides understanding of SFS vs. RS coherent structure in relationship to the effective LES filter cutoff within the Fourier description of scale, as determined by effective grid. This understanding will inform future analyses and a new modeling strategy for the RS-SFS contributions to the nonlinear terms in LES of flame-turbulence interactions centered on the development physically meaningful mathematical forms for scale-dependent SFS variables.

However, the three-dimensional chaotic forcings of a thin flame by the full range of turbulence eddy scales creates highly complex multiscale flame distortions in response to the wide ranges of eddy scales and strengths relative to the flame scale and flame strength local to a flame-eddy interaction. Spatial coherence resulting from these interactions is correspondingly highly complex, with coherent structure created over wide ranges of scale by the dynamics associated with the distorting flame. This complexity in multi-scale re-

sponse makes it difficult to dissect the Fourier-physical space relationships created by groupings of eddies at similar scale and strength. To reduce the levels of complexity from the impacts of a full range of 3D turbulence eddying motions on flame dynamics, and to more clearly describe coherent structure physical-Fourier space relationships in context with flame topography, we have created a “reduced-physics” model that captures a key underlying element in turbulence-flame dynamics: the distortion of a flame in response to repetitive forcing by single-scale eddies of controlled scale and strength relative to a well-defined flame sheet. Specifically, we design controllable non-chaotic interactions between an initially unstretched planar flame sheet and topologically well-defined eddies modeled as trains of two-dimensional single-scale vortices with controlled scale and strength relative to that of the flame. We argue that this flame-vortex interaction model captures essential deformations of a flame sheet in response to single-scale eddy motion. The reduction in dimensional and physical complexity produces well-defined Fourier-physical space relationships in coherent structure while retaining sufficient levels of complexity to apply the results extract key elements of turbulence-flame interactions that are local to flame-eddy interactions in premixed turbulent combustion.

With the reduced-physics flame-vortex interaction model we are able to choose the relationships between the flame scale/strength and eddy scale/strength in relationship to the interactions between a subset of eddies within the full range of turbulence-flame interactions that are ubiquitous in premixed turbulent combustion. As discussed in Sec. IIIB, the current study focuses on eddies with scale an order of magnitude larger than the characteristic flame scale. This ratio is chosen because turbulence eddies of order the flame scale tend not to strongly impact flame distortion²⁶, while the response to eddies larger in scale may be deduced from the current results. We argue that the current flame-eddy model characterizes local deformations of the flame sheet at the defined eddy scale with the degree of response determined by the strength of the defined single-scale eddies relative to the strength of the flame. In the complete series of studies underlying the current paper, relative eddy strengths are chosen to create “weak”, “moderate” and “strong” response in flame deformation. In the current paper we discuss in detail the “weak” response case, designed to create large repetitive distortions of the flame without pinching or the creation of pockets. The higher strength of the “moderate” case vortices was chosen to produce pinching (but no pockets), while the “strong” vortex case was designed to create repetitive pocket formation. Because the primary conclusions from this series of studies can be presented in context with “weak” case, we present detailed results for this case in the current paper. The “moderate” and “strong” cases are discussed in Sec. V in context with discussion of the application of the current analysis to 3D turbulence-flame interactions.

To apply the 2D Fourier description, the nonphysical spectral content from boundary discontinuities at boundaries is removed with the discontinuity pollution removal procedure described in Sec. II generalized to 3D signals (Appendix B and C). With 2D spectral filtering, we identify the relation-

ship between characteristic coherent features in physical and Fourier space associated with specific flame responses and we quantify the relative contributions of these features to the total variance content of key variables.

B. Overview of Numerical Simulation

In turbulent premixed flames, stiff nonlinear chemical reactions involving many reacting scalars introduce broad ranges of length and time scales that overlap with the characteristic scales of turbulence motions, resulting in strong two-way couplings between turbulence and combustion^{27–29}. These scales are typically organized in the form of key non-dimensional parameters within a parameter space that demarcates regimes of turbulent premixed combustion (Borghi²⁷, Peters²⁸, Williams²⁹).

The Karlovitz number is a key non-dimensional parameter that characterizes these regimes:

$$Ka \equiv \tau_F / \tau_\eta \sim \gamma (u_\eta / s_L)^2 \sim \gamma^{-1} (\delta_f / \eta)^2, \quad (12)$$

where $\tau_F = \delta_f / s_L$ is the characteristic flame time scale (s_L and δ_f are the laminar flame speed and thickness, respectively), $\tau_\eta = \eta / u_\eta = \eta^2 / \nu$ is the Kolmogorov time scale (u_η and η are the Kolmogorov velocity and length scales, respectively) and ν is the kinematic viscosity. In Eq. (12), $\gamma \equiv s_L \delta_f / \alpha \approx s_L \delta_f / \nu$, where α is the thermal diffusivity. $\alpha \approx \nu$ in air, even at high temperature and pressure. Assuming air, then, $\gamma \sim O(1)$ in the diffusion-dominated reaction zone. (In the inflow, where temperature and kinematic viscosity are an order of magnitude lower, $\gamma \sim O(10)$ or higher.) $Ka \ll 1$ is traditionally described as the “flamelet” regime while $1 \lesssim Ka \lesssim 10^2$ and $Ka \gtrsim 10^2$ are described as “thin”, and “broken” or “distributed” reaction zone regimes, respectively^{27–29}. Recent works^{26,30} suggest that the distributed combustion regime may be associated with higher $Ka \gtrsim O(10^4)$.

Some key fundamental dynamics of flame-turbulence interactions under distinct regimes may be conceptualized by reduced-physics systems that characterize local interactions between a premixed flame and single-scale turbulence eddies from within the inertial eddies within high Reynolds number turbulent reacting flow³¹, as illustrated experimentally in Filatyev *et al.*³². Using this framework, we represent the interaction between a laminar flame and single-scale inertial eddies of length scale r and velocity scale u_r with a train of 2D vortices with vortex scale $l_v \sim r$ and velocity scale $u_v \sim u_r$. To estimate Karlovitz number with vortex scales, we apply classical Kolmogorov 1941 scaling ($\varepsilon \sim u_v^3 / l_v$) to Eq. (12) to obtain:

$$Ka \sim (l_v / \delta_f)^{-1} (u_v / s_L) (Re_v)^{1/2}, \quad (13)$$

where $Re_v = u_v l_v / \nu$ is the vortex Reynolds number in the unburnt mixture and we model the vortices as characteristic inertial-range turbulence eddies.

In the numerical model, a vortex train was designed to characterize inertial-range eddies on the reactant side of high Ka turbulent premixed flames in the thin reaction zone regime³⁰

with integral scale Reynolds number $\sim 10^4$ and Karlovitz number ~ 1 , respectively. The vortex length scale was purposefully chosen to be an order of magnitude larger than the flame length scale ($l_v / \delta_f \approx 10$) to characterize the impacts of larger inertial range eddies and to better elucidate characteristics at disparate vortex and flame scales in Fourier space. The square of the vortex to flame velocity ratio, $(u_v / s_L)^2$, characterizes the ratio of vortex to flame strength. For the “weak” vortex case discussed in the current paper, $u_v / s_L \approx 0.5$.

As discussed in Sec. III A, whereas we discuss the key results in context with the weak case simulation, we also developed “moderate” and “strong” vortex simulations with $u_v / s_L \approx 2$, and 5, respectively. Whereas the weak case was designed to create strong repetitive distortions of the flame sheet, the moderate case was designed to create “pinching” distortions while the strong case was designed to create repetitive pinching and pocket formation. Most of the new knowledge from these studies was obtained with the weak case. As will be discussed in Sec. V, the strong case adds insight relevant to the application of these analysis to turbulence-flame interactions.

Table I summarizes relevant simulation parameters with the vortices at the location of the flame front in a corresponding cold-flow simulation. δ_f is defined as the distance between the iso-levels of 5% and 85% of the progress variable $C = (T - T_u) / (T_b - T_u)$, where T_u and T_b are the temperatures in the unburnt and burnt mixtures, respectively. The thermochemical conditions in the unburnt mixture (subscript u) shown in Table I are selected according to those used in the experiments of moderate Ka turbulent premixed flames by Skiba *et al.*³⁰.

Pressure	p	1 bar
Unburnt temperature	T_u	300 K
Unburnt equivalence ratio	ϕ_u	1
Laminar flame thickness	δ_f	0.5 mm
Laminar flame burning speed	s_L	39 cm/s
Vortex diameter	D	7.9 mm
Vortex length scale	l_v	5 mm
Vortex velocity scale	u_v	19 cm/s
Length scale ratio	l_v / δ_f	10
Velocity scale ratio	u_v / s_L	0.5
Vortex Reynolds number	Re_v	250
Karlovitz number	Ka	2.5

TABLE I: Important parameters for the flame-vortex interactions simulation.

The 2D numerical simulation was performed within the rectangular domain illustrated in Fig. 8, with domain lengths $L_y = 4D$ and $L_x = 16D$ in the cross-stream and streamwise directions, respectively, where D is the vortex diameter. At the initiation of the simulation, a pre-computed planar n -heptane/air laminar premixed flame was placed four vortex diameters downstream of the inflow boundary. An inflow of unburnt n -heptane/air mixture is injected into the domain in the streamwise direction x and the burnt products exit through the outflow plane. Periodic boundary conditions are applied in the cross-stream direction. An array of counter-rotating vor-

tex pairs is continuously injected, with specified mean inflow velocity, in the inflow boundary using time-varying Dirichlet boundary conditions for the velocity components. Convective outflow³³ is enforced in the outflow boundary.

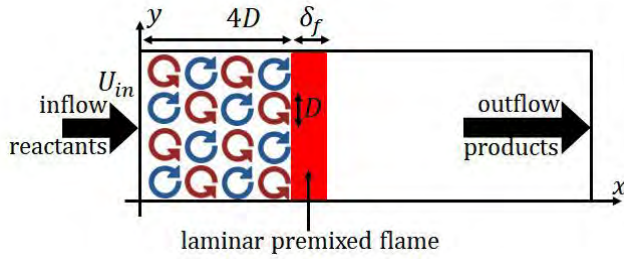


FIG. 8: Schematic of computational domain for the reduced physics 2D simulation of the interactions between a flame and an array of counter-rotating vortices.

The array of vortices is kinematically constructed from a combination of two Fourier modes (and their complex conjugates) as shown in 2D Fourier space in Figure 9b. The vector Fourier coefficients, perpendicular to the two primary wavevectors shown in 9b to satisfy incompressibility, are aligned in opposite directions. This combination produces an spatial array of single-scale counter-rotating vortices with circulation in opposite directions and with zero average linear or swirl velocity, as described in Brasseur and Wang⁵. Figure 9a shows a 2D isocontour of a normalized velocity component kinematically constructed using the combination of sine waves. A streamwise injection velocity U_{in} is added to the kinematic velocity field, where $U_{in} = 54$ cm/s to match the statistically averaged flame burning speed predicted in the simulation. In this way, the flame does not move, on average, in the statistically stationary limit. The flow is initialized with space-filling vortices on the reactants side of the computational domain ($x \leq 4D$).

The vortex length scale l_v characterizes the concentration of vorticity in each vortex core. We define l_v as the diameter of an equivalent circle with area equal to the area within the vortex that is bounded by the vorticity isocontour $0.3\omega_{peak}$, where ω_{peak} is the peak vorticity in the core. This area within the core captures more than 90% of the total vortex circulation. For the vortices of diameter D defined above, it turns out that $l_v = 0.63D$. The vortex velocity scale u_v , characterizing the strength of the vortex core, is defined as the circulation contained within the same vorticity isolevel divided by its perimeter.

The 2D flame-vortex interaction simulation was performed using the NGA code³³, where the unsteady compressible Navier-Stokes equations under the low Mach number and ideal gas approximations and the transport equations for temperature and mass fractions of 35 species are solved with boundary conditions. The scalar equations are discretized using the BQUICK scheme, which ensures that the physical bounds of species mass fractions are numerically preserved with minimal artificial diffusion³⁴. A recently-developed computationally-efficient, semi-implicit, iterative method is

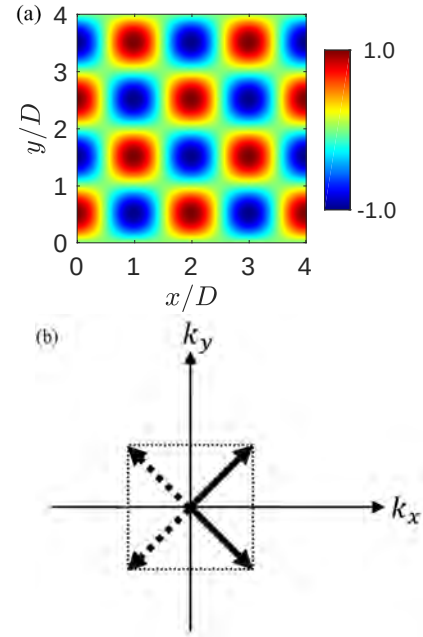


FIG. 9: Schematic of kinematically constructed velocity components for array of vortices injected in the inflow boundary. (a) Physical and (b) Fourier space view.

used for time-integration of the chemical source terms within the (gas-phase) species equations³⁵. This method has been shown to be as computationally efficient as an explicit time-integration per time step and allows for stable simulations with larger time steps than a corresponding explicit scheme³⁶.

A semi-detailed chemical kinetics model containing 35 species and 217 reactions (forward and backward reaction counted separately) is used³⁷, a mechanism reduced from the original 149 species and 1651 reactions³⁸ to reduce the computational cost of moderate Ka turbulent premixed flame simulations³⁷. Complete details with chemical model validation are given in^{37,39}.

The computational mesh is non-uniform in the streamwise direction and uniform in cross-stream direction. The grid is highly refined in the vortex-dominated reaction region and, especially, in the flame region in the streamwise direction and is stretched towards the outflow boundary. At least 80 grid points are used to resolve the vortex length scale in the reactants side and 20 grid points within the flame front thickness. A grid convergence study was carried out where we confirmed that the same results are obtained with twice as many grid points in both directions. The time step size is $1 \mu s$ to accurately resolve the chemical time scales.

C. Coherent Structure of Flame-Vortex Interactions in Physical Space

The numerical simulation is analyzed selecting four primary variables associated with key features of the 2D flame-vortex interactions: the streamwise components of momen-

tum vector (ρu and ρv) as representative of momentum impacts, the mixture enthalpy (ρh) containing thermal energy, and the formyl radical mass per unit volume (ρY_{HCO}) representing an intermediate species that identifies heat release rate by combustion⁴⁰. A single time instant within the statistically stationary state of the flame-vortex interactions is selected for analysis, when the flame is most “corrugated” as quantified by the maximum difference between the two extreme locations of the progress variable iso-level, $C = 0.5$. The statistically stationary state is observed after a transient period from the initial condition at approximately 4 vortex advection time scales, D/U_{in} .

Figure 10 shows the 2D instantaneous isocontours of primary variables, ρu , ρv , ρh , and ρY_{HCO} , over the region of $2 \leq x/D \leq 8$. Taken together and as a movie, Figures 10a-d show the interaction between the train of vortices and laminar flame at stationary state at peak corrugation. Note from Figures 10a and b that the vortices in the reactants that distort the flame are apparent in the momentum field but not in the energy or species. Interaction with the flame and baroclinic generation of vorticity at the flame lead to the development of elongated structures in the streamwise direction within the region downstream of the flame, impacted by flow acceleration and volumetric expansion within the flame front. As a consequence of baroclinic flame-generated vorticity^{41,42}, the vorticity within the elongated structures is of opposite sign to the vortices on the reactants side of the flame. It is for this reason that ρv changes sign across the flame (Fig. 10b).

As vortices are continuously injected into the domain from the inflow boundary, the flame is perturbed periodically in time (not shown here) and symmetrically in y . The train of vortices causes the formation of flame cusps when the induced velocity of two adjacent counter-rotating vortices locally advects the flame sheet into the burnt products between the vortices (at $y/D = 1$ and 3 in Fig. 10). The strength of the vortices relative to the flame sheet determines the extent to which the cusp regions extend into the products. In contrast with momentum components, ρh and ρY_{HCO} in Fig. 10 are approximately spatially uniform on the reactants and products sides of the flame with large gradients localized to the thin flame. ρh has a jump from reactants to products with local scale on order the laminar flame thickness, δ_f . ρY_{HCO} , by contrast, is a combustion intermediate that is highly localized with a strong peak within the reaction zone internal to the flame at a scale roughly three times smaller than the laminar flame thickness, δ_f . The intermediate species generated and destroyed within the reaction zone and that contribute strongly to heat release are of this type.

From Fig. 10, four distinct coherent structural features may be identified in physical-space in the primary variables: (1) vortices in momentum on the reactants side, (2) elongated structures in momentum on the products side (we shall refer to these as “streaks”), (3) a corrugated flame front with locally large gradients in all variables, particularly in thermal energy and species concentration, and (4) strong streamwise jumps in energy, temperature, and some species such as fuel and oxidant (not shown) between reactants and products. In the next sections, these key physical-space coherent structural features

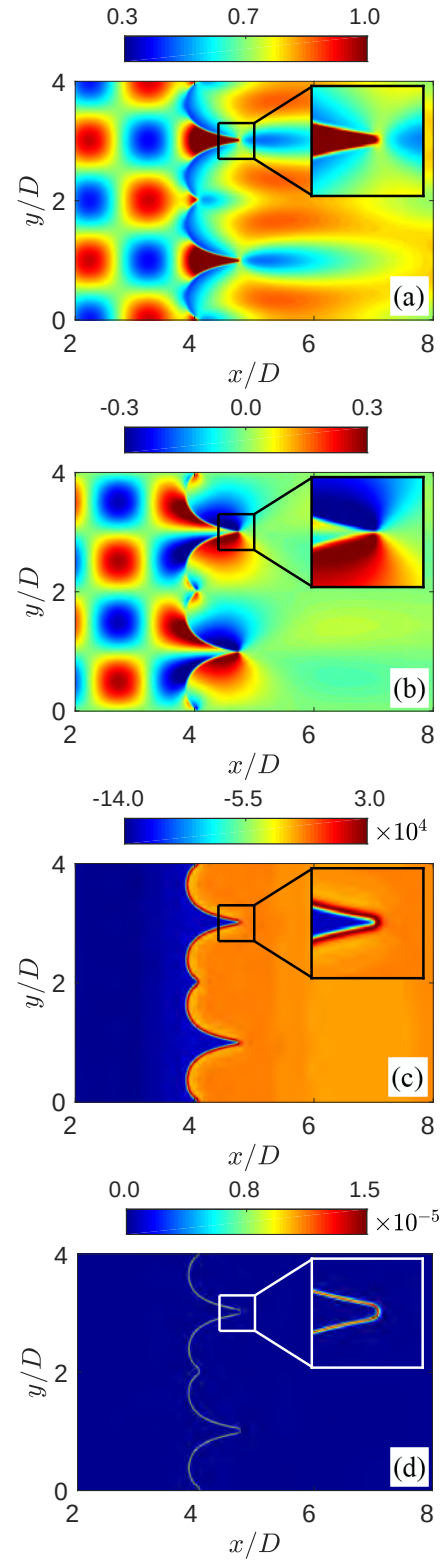


FIG. 10: Isocontours of primary variables in physical space for the flame-vortex interaction simulation. (a) ρu in $\text{kg}/\text{m}^2\text{s}$; (b) ρv in $\text{kg}/\text{m}^2\text{s}$; (c) ρh in J/m^3 ; (d) ρY_{HCO} in kg/m^3 .

are identified with coherent features in Fourier space and are

discussed in relationship to scale-decomposition within the classical LES framework.

To apply the multidimensional Fourier description to the 2D nonperiodic signals described, the nonphysical spectral content that arises from discontinuities at the boundaries in x must be removed using the “discontinuity pollution removal” procedure described in Sec. II C for 1D signals but extended to 2D. In Appendix B we describe the extension to 3D signals with one nonperiodic direction; we apply this method reduced to 2D. In Appendix C the multidimensional procedure was validated with $\alpha = 0.85$, which ensures that the maximum signal modification remains below 5%. As a result, the vortices in the reactants ($x/D < 4$), elongated streaks in the products ($x/D > 4$), and large gradients within the flame front ($x/D \approx 4$) observed in Fig. 10 are virtually unchanged in the pollution removal periodization process (see Fig. 25 in Appendix C).

IV. FOURIER-PHYSICAL SPACE COHERENT STRUCTURE OF FLAME-VORTEX INTERACTIONS

Figure 11 presents the concurrent Fourier space view of the physical-space signals shown in Fig. 10 described in Sec. III C. In Fig. 11 we plot isocontours of the logarithmic squared Fourier coefficients for ρu , ρh , and ρY_{HCO} . The wavenumber axes are normalized using the laminar flame thickness, the scale defined by the boundaries of the domain in Fig. 11. The pink box identifies the vortex scale ($k_{x,y}\delta_f/\pi = \pm\delta_f/D = 0.06$) and is associated with the four Fourier modes used to construct the train of vortices that enter the computational domain (Fig. 9).

In Fourier space, the squared Fourier coefficients are symmetric due to the reality condition. An additional symmetry arises in k_y because the array of symmetrical vortices create a symmetrical response in this laminar flow. Thus, $\widehat{\phi}^2(k_x, k_y) \approx \widehat{\phi}^2(k_x, -k_y)$. The forced even spanwise symmetry in y leads to negligible Fourier content in Fourier modes with wavenumber $k_y/\Delta k_y = \pm 1, \pm 3, \dots$. To visualize better the structure, the odd k_y modes are removed from all isocontour plots. We refer to “Fourier energy content” as the squared Fourier coefficients, which represent wavenumber-specific contributions to variance. To better understand how coherent structure (described in Sec. III C) is characterized in Fourier-space, systematic spectral filtering is applied.

A. Analysis of Small-Scale Structure

For the primary variables shown in Fig. 11, the highest magnitude Fourier energy content is found to be organized within a star-like pattern towards high wavenumbers (i.e. small scales) that extend from the vortex to flame scale. ρY_{HCO} energetic Fourier content extends to $k_{x,y}\delta_f/\pi \approx \pm 3$ (not shown here), the reaction zone scale within the premixed flame.

To identify the physical-space content associated with the correlated collections of Fourier modes within the legs of the

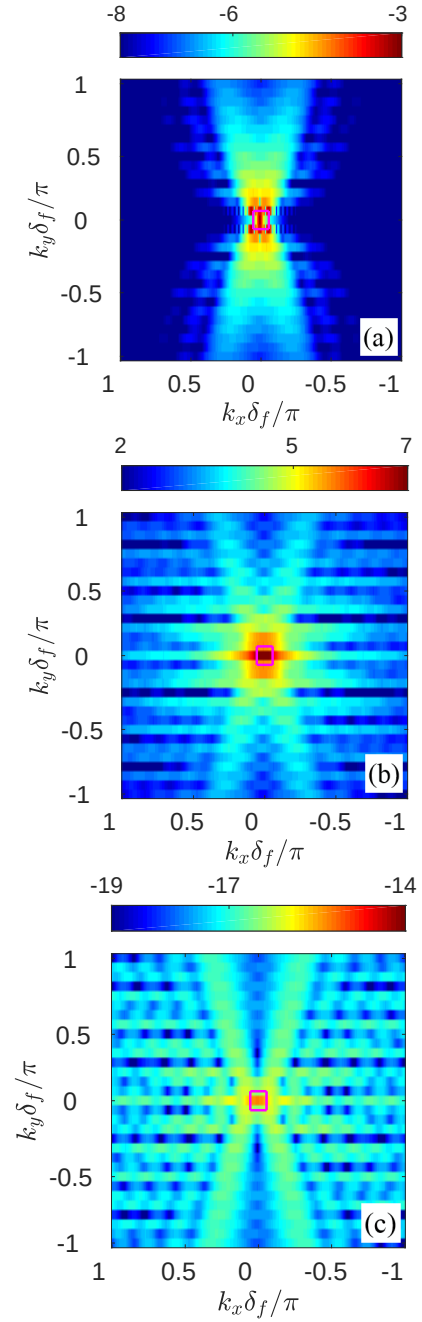


FIG. 11: Isocontours of the logarithmic squared Fourier coefficients of the same primary variables at the same time instant in flame-vortex interaction as Fig. 10. The pink box identifies the vortex scale ($k_{x,y}\delta_f/\pi = \pm\delta_f/D = 0.06$). (a) $\log_{10}(\widehat{\rho u}^2)$; (b) $\log_{10}(\widehat{\rho h}^2)$; (c) $\log_{10}(\widehat{\rho Y_{HCO}}^2)$.

star-like pattern in 2D spectral content in Fig. 11, we designed localized elliptical filters (conceptually band-pass-like) that we optimally aligned with the legs of the star patterns and their symmetric counterpart. The two characteristic scales of the ellipse in 2D Fourier space were estimated qualitatively. In Fig. 12 we show the filtered signals above and the unfiltered signals below for momentum, energy, and species

concentration variables shown in Figs. 10 and 11. Whereas the filter is designed to capture the four dominant legs in the star-like pattern, the wavenumbers within the vortex-scale box ($|k_{x,y}|\delta_f/\pi \leq 0.06$) are not captured by the filter. Indeed, the legs of the star-like pattern extend from the vortex scale box to, and beyond, the flame scale box.

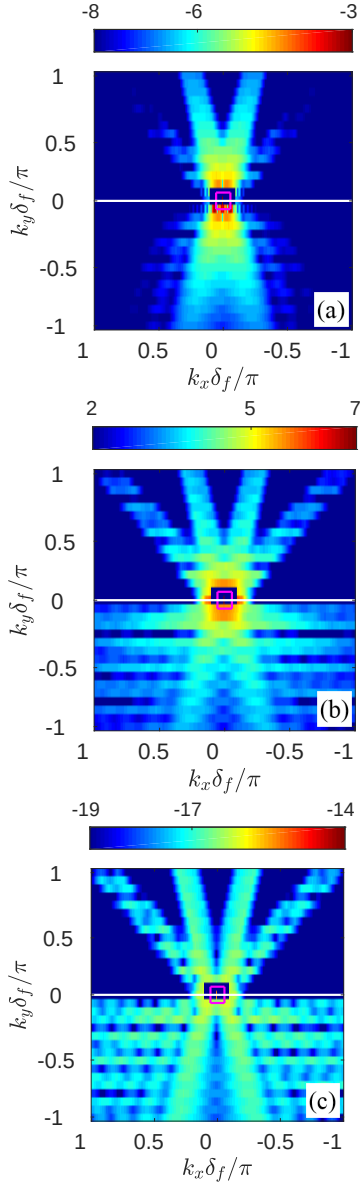


FIG. 12: Isocontours of the logarithmic squared Fourier coefficients of primary variables shown in Fig. 11. The filtered and original signals are shown in the top and bottom halves of each figure, respectively. As discussed in the text, the filter is constructed to extract Fourier content within the star-like pattern in Fourier space and the pink box identifies the vortex scale ($k_{x,y}\delta_f/\pi = \pm 0.06$). (a) $\log_{10}(\widehat{\rho u}^2)$; (b) $\log_{10}(\widehat{\rho h}^2)$; (c) $\log_{10}(\widehat{\rho Y_{HCO}}^2)$.

Figure 13 shows in physical space the filtered and original signals in the top and bottom halves of each isocontour,

respectively. The mean value of the variable was removed by the filter, so the mean values are added to the filtered signals in Fig. 13 for direct comparison with the original physical-space signals. We observe that the star-like region of Fourier space corresponds to physical-space structures that surround and define the corrugated flame front. The smaller scale features of the flame front in the insets of Fig. 10 are captured by the coherent star-like regions of Fourier space identified in the insets of Fig. 13. In particular, these regions of Fourier space appear to recover most of the flame cusps in ρu . However, not all the corrugations of the flame are recovered, as observed by the sharp angles between segments of filtered ρY_{HCO} in Fig. 13c compared with the smooth large-scale segments in the original signal.

As discussed in Sec. II, the Fourier variance content of a discontinuity has power-law decay to the grid cutoff scale. The star-like patterns in Fourier space are, in reality, power-law decays arising from near-discontinuities at the flame front. Given the finite thickness of the flame front and the sharp, but smooth, variation across the front, the Fourier energy content exhibit power-law decay down to the flame front scale. At the flame thickness, the Fourier variance transitions to exponential decay (to the grid cutoff scale). The directions of the “star legs” are normal to the corresponding sections of flame front that generates the power-law decay.

To better understand the connection between the star-like pattern in Fourier space and the corrugated flame front in physical space, we compute the flame-normal gradient of primary variables along the flame centerline (progress variable iso-level, $C = 0.5$). The unit normal vector direction is defined as $\vec{n} = -\nabla T/|\nabla T|$ and indicated as $\vec{n}_f$ at the flame centerline.

Figure 14 shows the position of the flame centerline iso-level in the (x, y) plane colored according to the magnitude of local flame-normal gradient of each variable multiplied by the laminar flame thickness, which provides an estimate of the local “jump” across the flame. We note that the normal gradients are highly localized to the flame sheet and that, whereas the magnitude of the local normal gradient is relatively uniform along the sheet with ρh and ρY_{HCO} concentration, this is not the case with momentum, which shows large variability along the sheet.

Figure 15 shows the Probability Density Function (PDF) of the angle θ_f between the flame centerline direction $\vec{n}_f$ and the streamwise direction, x (counter clockwise direction). This PDF is approximately symmetric, as the flame corrugations are caused by a symmetric array of vortices along the cross-stream direction. Four dominant peaks around $\pm 45^\circ$ and $\pm 75^\circ$ may be identified, which match the angles of dominant legs in the star-like pattern observed in Fig. 12. The dominant peaks in the PDF at $\theta_f \approx \pm 75^\circ$ are associated with the more pinched flame cusps ($y/D = 1$ and 3 in Fig. 13), which cover a large segment of the flame centerline, and the secondary peaks at $\theta_f \approx \pm 45^\circ$ represent the smaller flame cusps ($y/D = 0, 2$, and 4 in Fig. 13).

The larger magnitude of the flame-normal jump of ρu over the more pinched flame cusps (Fig. 14a) produces Fourier content dominantly along the legs oriented at $\pm 75^\circ$ (Fig. 12a). In contrast, both thermochemical variables are shown to have

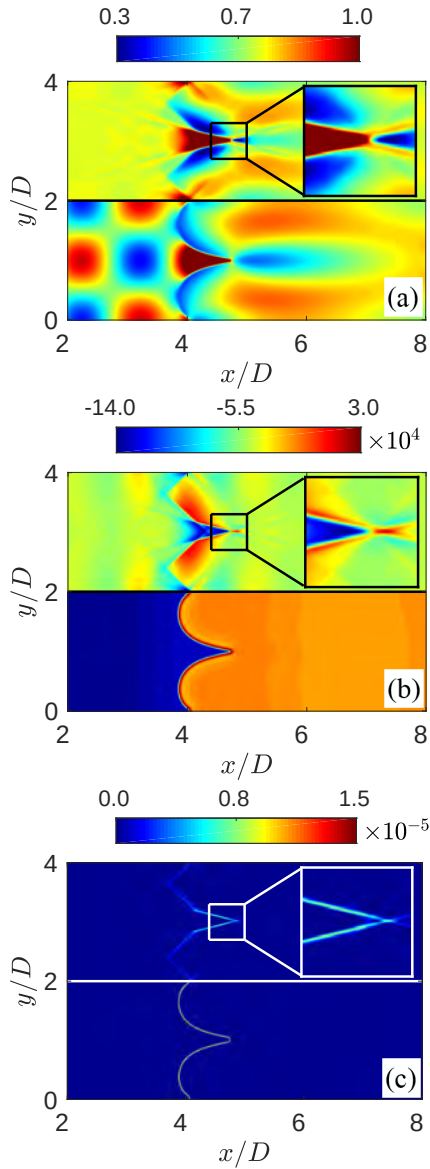


FIG. 13: Isocontours of primary variables in physical space shown in Fig. 10. The filtered and original signals are shown in the top and bottom halves of each isocontour, respectively.

As discussed in text, filter is constructed to extract Fourier content within the star-like pattern in Fig. 12. Insets show the filtered signal at the flame cusps. (a) ρu ; (b) ρh ; (c) ρY_{HCO} .

approximately uniform flame-normal jump over the flame centerline (Fig. 14b-c), as the internal flame structure is preserved during the relatively weak ($u_v/s_L \approx 0.5$) flame-vortex interactions.

The variability in the magnitude of the flame-normal jump along the flame centerline is quantified by its intensity (standard deviation divided by mean). The intensities of ρu , ρh , and ρY_{HCO} are 84%, 6.8%, and 9.1%, respectively. Thus, the star-like pattern of ρh and ρY_{HCO} is approximately insensitive to the magnitude of flame-normal jump and is dictated by the PDF of θ_f . Therefore, dominant legs in the star-like pat-

tern of ρh and ρY_{HCO} are observed at $\pm 75^\circ$, secondary legs at $\pm 45^\circ$, and non-negligible content over a broad range of angles (Fig. 11b-c).

The analysis of this section indicates that physical-space coherent structure associated with the corrugated flame front creates organization (coherence) in Fourier space that, at the smaller scales, correspond to a star-like pattern with significant Fourier energy content extending from the vortex scale box to the flame scale and smaller (Figs. 12 and 13). From the perspective of typical LES scale decomposition, this physical-Fourier space duality in coherent structure would be partially resolved by the grid and dynamics, and partially subfilter-scale. The understanding of dynamically coherent structure could be impactful to dynamically relevant SFS modeling. In the next section, we define RS consistent with the LES framework to facilitate identification and quantification of coherent structure in the physical-Fourier space duality at the RS and SFS consistent with classical LES.

B. Defining Resolved vs Subfilter Scales Consistent with LES

In the classical LES framework, resolved fluctuations defined by the effective grid are carried forward in time in the numerical simulation, where “effective” implies the additional coarsening of the geometric grid due to the impacts of the numerical and model dissipation. A geometric grid properly designed for LES is one in which the corresponding effective grid resolve a large percentage of variance of fluctuations in momentum and energy where “large” implies capturing covariances at the 80-90% level. As discussed below, here we require at least 85% of the resolved variance of momentum ($\rho \vec{u}$) and enthalpy (ρh) per unit volume. Variance is calculated after removing two-dimensional averaged values, equivalent to the sum of squared Fourier coefficients excluding $\vec{k} = 0$.

We define “resolved scales” using a 2D sharp low-pass rectangular “effective grid” filter in Fourier space with wavenumber cutoffs $k_{G,x}$ and $k_{G,y}$. We define a single characteristic filter wavenumber scale as $k_G \equiv \sqrt{k_{G,x}k_{G,y}}$. The optimal wavenumber cutoffs ($k_{G,x}, k_{G,y}$) that define the RS wavenumber cutoffs are obtained performing parametric variations to minimize k_G with specified fixed percentage of total captured variance over the statistically stationary regime of the simulation. Figure 16 presents 2D isocontours of k_G over the $k_{G,x} - k_{G,y}$ parameter space nondimensionalized by the laminar flame thickness. The colored iso-lines in Fig. 16 indicate wavenumber cutoffs ($k_{G,x}, k_{G,y}$) that capture at least 70%, 80%, 85%, and 90% of the total variance for ρu , ρv , and ρh . The solid circles identify the corresponding optimal wavenumber cutoffs based on minimum k_G for each iso-line, which provides the respective wavenumbers $k_{RS,x}$ and $k_{RS,y}$ that defined the rectangular RS region in 2D Fourier space.

Figure 16 indicates that the RS wavenumber cutoffs are systematically asymmetric $k_{RS,y} > k_{RS,x}$. This is due to the cross-stream corrugations of the flame front with content at higher k_y . The observation that the colored iso-lines in Fig. 16 become approximately vertical and horizontal at higher $k_{G,y}$ and

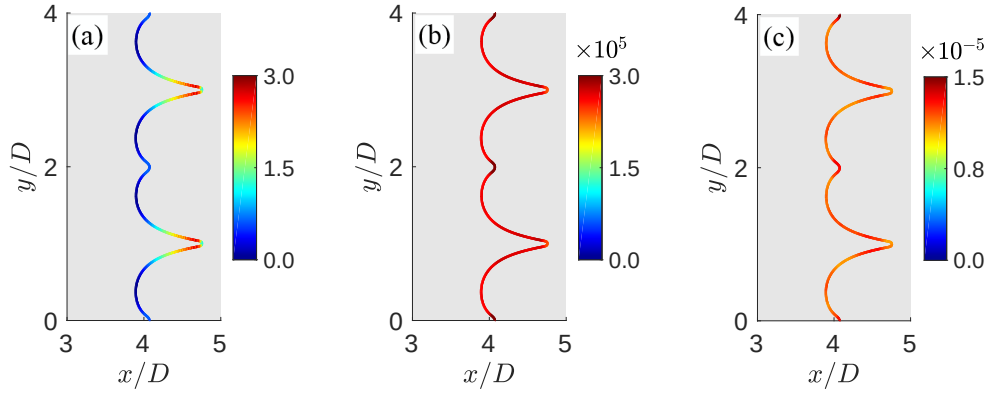


FIG. 14: Magnitudes of flame-normal gradients (a) $\delta_f |\vec{n}_f \cdot \nabla(\rho u)|$ ($\text{kg}/\text{m}^2\text{s}$), (b) $\delta_f |\vec{n}_f \cdot \nabla(dph)|$ (J/m^3), and (c) $\delta_f |\vec{n}_f \cdot \nabla(\rho Y_{HCO})|$ (kg/m^3) over the flame centerline defined by progress variable iso-level, $C = 0.5$.

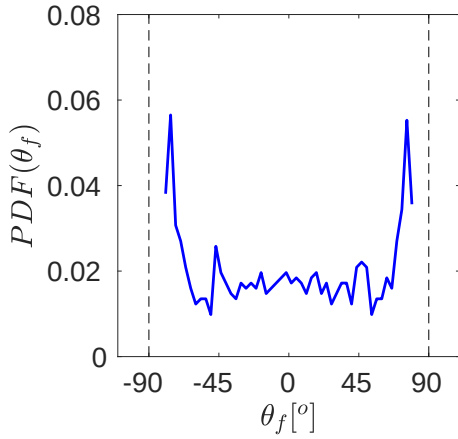


FIG. 15: PDF of local flame centerline angle θ_f between the flame centerline direction $\vec{n}_f$ and the streamwise direction (counter clockwise direction).

$k_{G,x}$ reflects the rapid drop in variance that occurs as the filter cutoff crosses the wavenumbers associated with the vortices, $k_{G,(x,y)}\delta_f/\pi = 0.06$, which contain a large percent of momentum variance. ρh variance is largely contained in first few wavenumbers in 2D Fourier space (Fig. 11b), so the colored iso-lines shown in Fig. 16 solely depend on captured momentum variance. The conclusions from this study were found to be qualitatively insensitive to the choice for percentage variance in RS fluctuations between approximately 80 to 90%. We therefore chose *at least* 85% variance in momentum and enthalpy per unit volume to define “resolved scales”. The resulting wavenumber cutoffs are $k_{RS,x}\delta_f/\pi = 0.1$ and $k_{RS,y}\delta_f/\pi = 0.26$ (yellow symbol in Fig. 16). This corresponds to the smallest resolved lengths scale in the streamwise and cross-stream directions $\ell_{RS,x} = \pi/k_{RS,x} = 0.62D$ and $\ell_{RS,y} = \pi/k_{RS,y} = 0.24D$, respectively.

The procedure described above does not include the variance content of ρY_{HCO} , which as previously discussed characterizes heat release in the reaction zone that is generally unresolved in practical LES. In the current analysis, for exam-

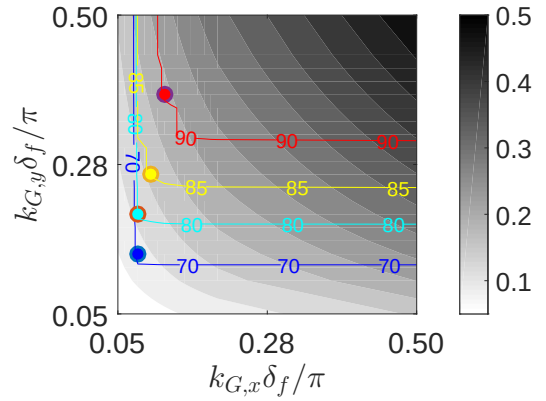


FIG. 16: Isocontours of nondimensional filter wavenumber cutoff $k_G \delta_f / \pi$ over the $k_{G,x} - k_{G,y}$ parameter space. Colored iso-lines indicate percentage variance content captured by the filter relative to the energy content of the original signal. Solid circles indicate the minimum k_G at given percentage variance content level.

ple, the wavenumber cutoffs would increase to $k_{G,x}\delta_f/\pi = k_{G,y}\delta_f/\pi = 3$ to capture 85% of the variance of ρY_{HCO} , which would result in ρu and ρh being nearly fully resolved by the effective grid. In this case, the simulation would be close to a DNS. We conclude that not all dynamical equations for turbulent reacting flows can be treated within the classical LES framework and SFS modeling strategies. This is because key chemistry-related dynamics necessarily takes place at scales associated with molecular diffusion which is not resolvable in practical LES, invalidating the key LES assumption that all dominant dynamics is captured by the effective grid and that the models for SFS terms must primarily capture forward-cascade dynamics.

C. Analysis of Resolved-Scale Structure

We continue our analysis within the LES framework using the effective grid filter described in Sec. IV B. Figure

17 shows in physical space the RS and original signals in the top and bottom halves of each isocontour plot, respectively. In these figures we show the RS flame front with boundaries defined by isocontour levels 0.05 and 0.85 of the resolved temperature-based progress variable $C^r = (T^r - T_u^r)/(T_b^r - T_u^r)$, where superscript r indicates RS variable. Figure 17 indicates that the large scale structural features of key variables are readily identified in the full field (bottom halves of each isocontour). These include the vortices in the reactants side and the elongated streaks on the products side for ρu (Fig. 17a). The RS corrugated flame front is less corrugated and thicker, with corresponding lower flame-normal gradients within the front, than the full field which contains the small-scale features that exist within the SFS motions.

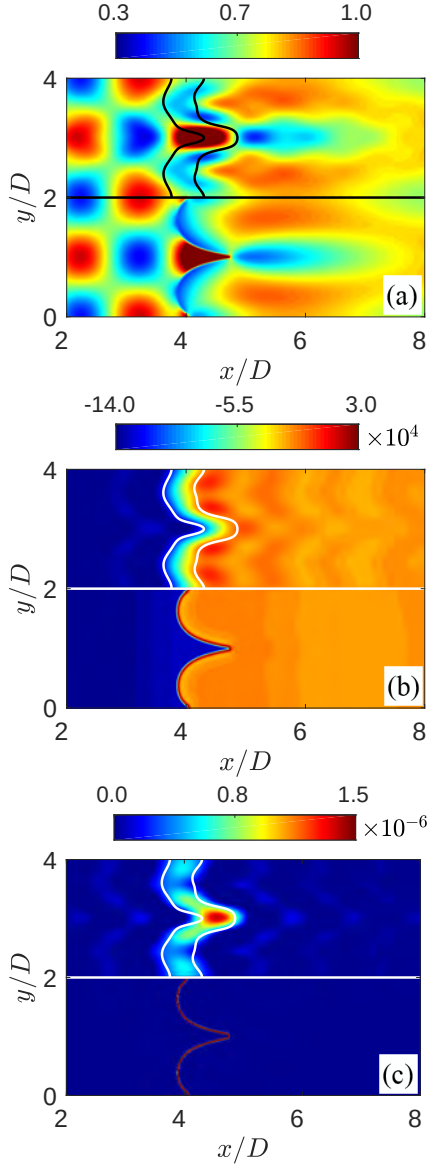


FIG. 17: Isocontours of primary variables in physical space.

Resolved and original signals are shown in the top and bottom halves of each isocontour plot, respectively. (a) ρu in kg/m^2s ; (b) ρh in J/m^3 ; (c) ρY_{HCO} in kg/m^3 .

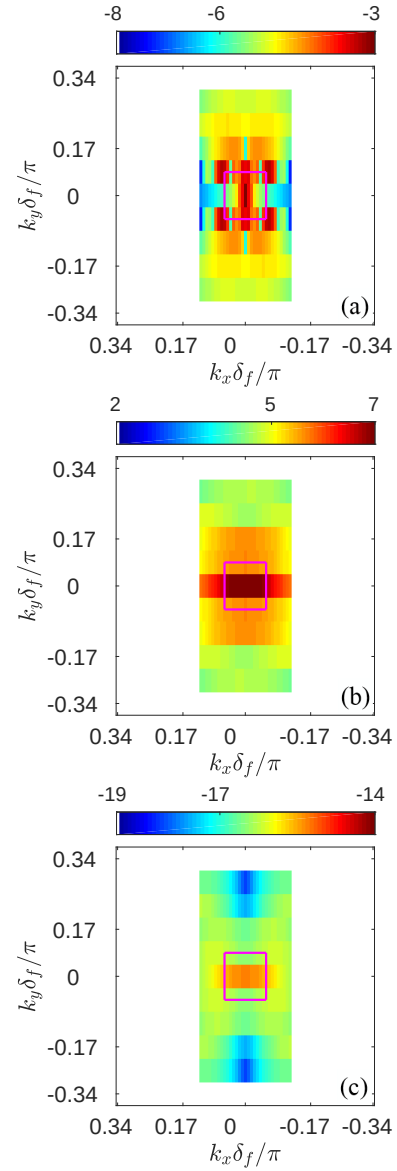


FIG. 18: Isocontours of the logarithmic squared Fourier coefficients of primary variables at the resolved scales. SFS modes are in the white at the figures. (a) $\log_{10}(\widehat{\rho u}^2)$; (b) $\log_{10}(\widehat{\rho h}^2)$; (c) $\log_{10}(\widehat{\rho Y_{HCO}}^2)$.

Figures 12 and 13 describe coherent Fourier and physical space features at the flame front that are associated with the star-like pattern that extend from the low wavenumber vortex scales (magenta box in Figs. 12 and 13) to the high wavenumbers at the flame and reaction-zone scales (beyond the domain boundaries in Figs. 12 and 13). However, accurate LES requires accurate prediction of RS dynamics. Thus, the relationships between RS coherent structure in Fourier vs. physical space are of primary interest. Figure 18 presents the concurrent view in Fourier space of the RS signals show in the top halves of Fig. 17, where isocontours of the logarithmic squared Fourier coefficients for each primary variable are shown within the Fourier-space rectangle defined by

our determined RS wavenumber cutoffs ($|k_x|\delta_f/\pi \leq 0.1$ and $|k_y|\delta_f/\pi \leq 0.26$). The color bars of these figures span the same range as in Fig. 12 and the magenta lines correspondingly indicate the vortex-scale box ($k_{x,y}\delta_f/\pi = \pm\delta_f/D = \pm 0.06$). SFS modes are identified by the white regions in Fig. 18. In contrast to the star-like pattern discussed in Sec. IV A, which covers a wide broadband range of mostly SFS fluctuations, the Fourier content towards low wavenumbers (i.e. larger RS motions) is shown to be localized over a reduced number of Fourier modes, such as the four corners of the vortex scale box in ρu in Fig. 18a.

By systematically filtering such spectral regions of interest within the RS rectangle in Fig. 18 using localized rectangular sharp filters, we have identified four distinct regions in Fourier space in Fig. 19 that characterize distinct features of the resolved scales. We systematically compute the inverse Fourier transform of the filtered Fourier coefficients within the four sub-regions identified in Fig. 19 and analyze the corresponding physical-space structure. The distinct colored subregions in Fig. 19 represent the 2D sharp Fourier space filters employed to decompose the resolved scales.

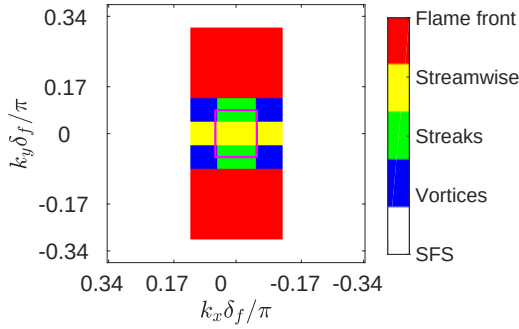


FIG. 19: Four distinct color-coded RS sub-regions in Fourier space.

1. Vortices

Figure 18a shows high magnitude Fourier content at and near the corners of the vortex-scale box for ρu . The four corners are, by construction, the modes used to generate the single-scale vortices injected into the flow through Dirichlet boundary conditions at the inflow boundary (Fig. 9). The Fourier coefficients surrounding these forcing modes are isolated using the 2D sharp Fourier space filter identified by the blue regions of Fig. 19, which filters Fourier modes in the spectral areas $|k_y|\delta_f/\pi = 0.06$ and $0.05 < |k_x|\delta_f/\pi \leq k_{RS,x}\delta_f/\pi$. Figure 20 shows the physical space view of the sub-filtered field associated with RS vortices above in comparison with the complete RS field below. The filtered ρu field characterizes the vortices on the reactants side of the mixture, effectively matching the complete RS signal up to $x \lesssim 3.5D$. If only the four Fourier coefficients associated with the corners of the vortex-scale box were extracted, the filtered physical-space signal would describe vortices spread

uniformly over the entire computational domain. The additional Fourier modes and their phase relationships suppress the signal associated with the vortices on the products side of the flame, localizing the vortices in the flame-vortex interactions to the reactants side of the mixture.

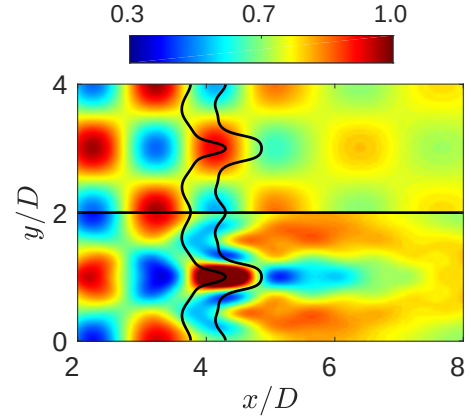


FIG. 20: Above we show the physical space view of the sub-filtered field associated with the RS vortices (blue subregions of Fig. 19) in comparison with the complete RS field shown below for ρu (kg/m^2s).

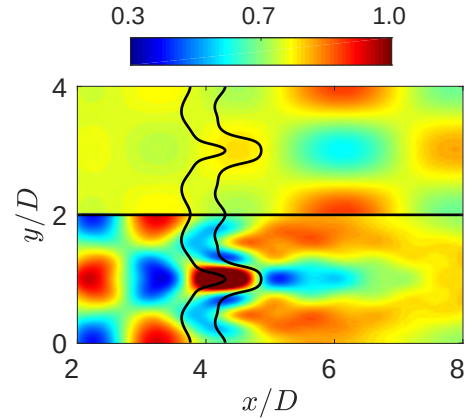


FIG. 21: Above we show the physical space view of the sub-filtered field associated with the RS elongated streaks (green subregions of Fig. 19) in comparison with the complete RS field shown below for ρu (kg/m^2s).

2. Elongated Streaks

The high magnitude RS Fourier content observed for ρu in Fig. 18a and identified by the green regions in Fig. 19 at k_x surrounding zero and $k_y\delta_f/\pi = \pm 0.06$, is associated with physical-space structure that has characteristic length scales in the streamwise direction x that are larger than the vortex scales in y where $k_y\delta_f/\pi$ is that for vortices created to force the flame. The Fourier coefficients isolated using 2D sharp

Fourier space filter over the green region in Fig. 19 are located at $|k_y|\delta_f/\pi = 0.06$ and within $|k_x|\delta_f/\pi \leq 0.05$. Figure 21 compares the inverse FTF of the sub-filtered ρu field above with the complete RS field below. The sub-filtered RS field characterizes the elongated streaks in the burned products side of the mixture. The degree of localization of the filtered physical-space signal towards the burnt products side is a result of the phase correlations among the Fourier coefficients within the green sub-filtered region in Fig. 19. The relationship between the Fourier coefficients associated with vortices and elongated streaks is controlled by the dynamical interaction between the vortices and flame kinetics to create elongated streaks due to volumetric expansion as the vortices interact with the flame front.

3. Corrugated Flame Front

As discussed in Sec. IV A, the physical-space coherent structure associated with the highest gradients associated with the reaction zone in the corrugated flame front is organized in Fourier space within a star-like pattern that extends from the vortex scale box to beyond the flame scale box (Fig. 12), that is from the smallest SFS motions to within the RS motions. Thus, in an LES the physical-space structure associated with these Fourier coefficients are partially resolved scale and partially subfilter scale. The RS content of the corrugated flame is characterized by the Fourier modes within the red spectral “flame front” region in Fig. 19, the region $0.06 \leq |k_y|\delta_f/\pi \leq k_{RS,y}\delta_f/\pi$ and $|k_x| \leq k_{RS,x}$. The sub-filtered physical-space counterpart of these sub-filtered Fourier coefficients is presented in Fig. 22 for primary variables of interest in the upper half of each figure for comparison with their respective complete RS signals in the lower half of each figure. Figure 22 shows that the sub-filtered signals characterize, in physical space, the RS flame cusp associated with the RS flame front corrugations in the cross-stream direction. The black lines in Fig. 22 indicate progress variable values 0.05 and 0.85 in the RS temperature field. The variance associated with these Fourier coefficients makes the RS wavenumber cutoffs systematically non-isotropic ($k_{RS,y} > k_{RS,x}$, in Sec. IV B), consistent with the RS structure of the large-scale cross-stream corrugations in the flame front. Therefore, the largest scale content of the star-like pattern described in Sec. IV A (Fig. 12) is resolved by the LES effective grid, with the higher gradient small-scale features of the corrugated flame mostly subfilter scale.

4. Streamwise Front

The final physical/Fourier-space coherent structure identified is associated with the cross-stream averaged gradients in the corrugated flame front, referred as “streamwise front” in Fig. 19, characterized by the Fourier modes within the yellow spectral sub-region, the region $k_y = 0$ and $|k_x| \leq k_{RS,x}$. Figure 23 shows the physical space view of the sub-filtered field associated with streamwise gradients in the flame front in com-

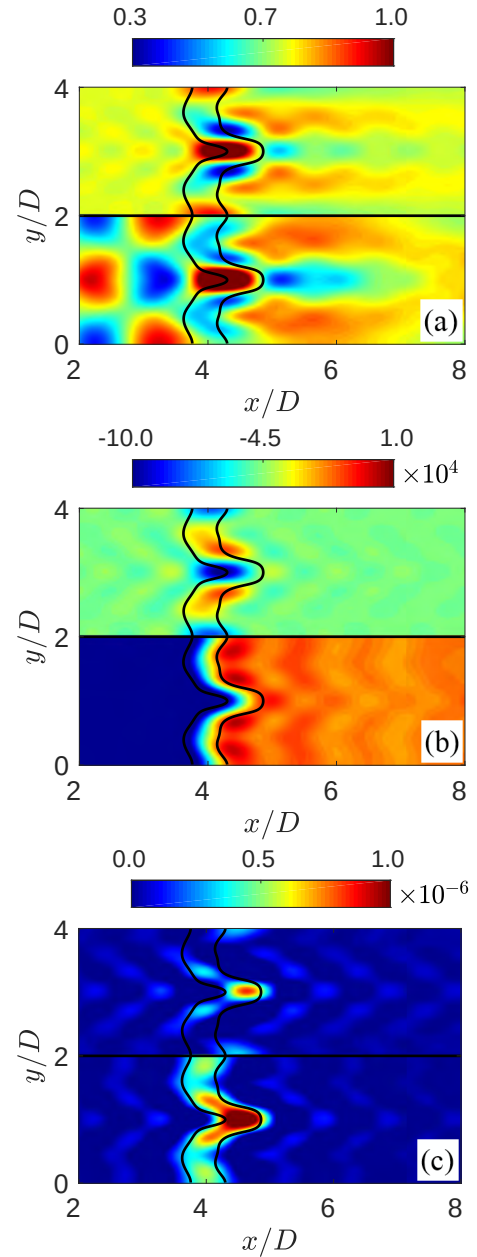


FIG. 22: Above we show the physical space view of the sub-filtered field associated with the RS corrugated flame front (red subregions of Fig. 19) in comparison with the complete RS field shown below for. (a) ρu ($\text{kg}/\text{m}^2\text{s}$); (b) ρh (J/m^3); (c) ρY_{HCO} (kg/m^3).

parison with the complete RS field (below). In particular, the streamwise flame front reflects the jump of ρh from reactants to products and the localized concentration of ρY_{HCO} within the flame front, as described in Sec. III C.

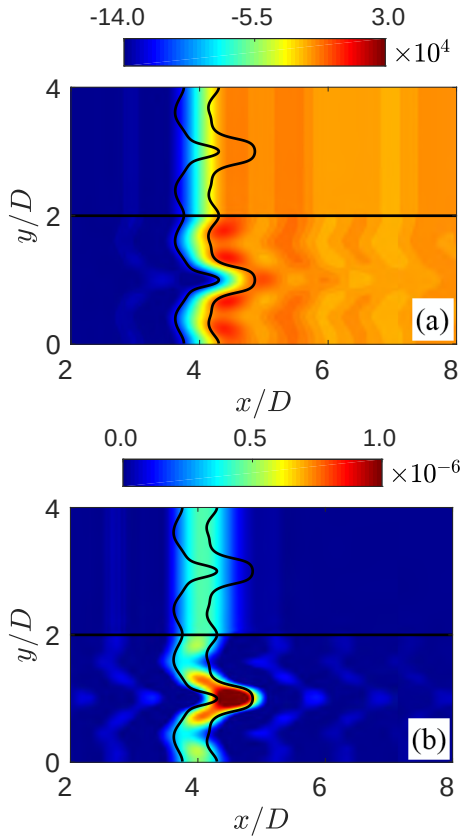


FIG. 23: Above we show the physical space view of the sub-filtered field associated with the RS streamwise front (yellow subregions of Fig. 19) in comparison with the complete RS field shown below for. (a) ρh (J/m^3); (b) ρY_{HCO} (kg/m^3).

D. Energy Content in Distinct Structures

The systematic filtering operations described in Sec. IV C relate coherent structure in physical space to coherent structure in Fourier space in context with LES of premixed turbulent combustion. To quantify the relative variance content of the distinct physical/Fourier-space coherent structures for primary variables of interest, we compute the filtered variance content of each coherent structure as the sum of squared Fourier coefficients over the sub-regions in RS Fourier space shown in Fig. 19. These, we have found, characterize distinct structural features in physical space. Figure 24 presents for each variable the percentage of variance contained in each coherent structure relative to the energy contained in the total original signal.

The sum of the color bars in Fig. 24 gives the total % variance resolved by the effective grid. We note that momentum and enthalpy are resolved, by construction, at least at the 85% level. Variance of ρY_{HCO} , in the other hand, is only 8% resolved. Created and destroyed in the reaction zone, HCO as of several important species with dynamics underlying heat release largely unresolved by practical LES. As discussed in Sec. IV A, the star-like pattern is associated with reaction-

zone kinetics. This narrow frontal structure exists within the corrugated RS flame front described in Sec. IV C and is the dominant SFS coherent structure.

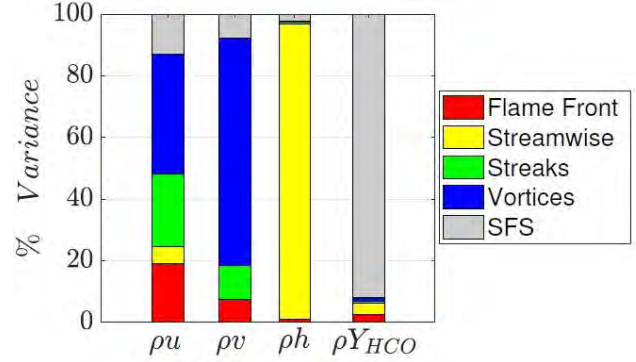


FIG. 24: Percent variance content in ρu , ρh , and ρY_{HCO} in RS flame front (red), RS streamwise front (yellow), RS elongated streaks (green), RS vortices (blue), and SFS motions (gray) for the flame-vortex interaction simulation. Percent variance is relative to total variance of the complete (RS and SFS) signal.

Figure 24 indicates that the RS variance of ρu is dominated by the vortices, elongated streaks, and RS corrugated flame, with relatively small contribution from the streamwise front. In contrast, the RS variance of ρh is fully dominated by the streamwise front.

On the other hand, the dominantly SFS variance of the species ρY_{HCO} is associated with the internal reaction-zone structure of the flame where heat release is highly localized. These results suggest that within a classical LES framework with an effective grid designed to capture a large percentage of integral-scale momentum fluctuations in a turbulent premixed flame, evolves turbulence eddies in momentum not only in the reactants and products (vortices and elongated streaks), but also in the RS flame front. Since the corrugated flame front is a coherent structure that resides partially at resolved and subfilter scales, one would expect dominant inter-scale couplings exist among Fourier modes associated with this coherent structure. The modeling of inter-scale couplings are more critical for the combustion intermediates that are directly involved in heat release, such as ρY_{HCO} , where variance content is largely at the subfilter scales. The turbulent combustion dynamical system therefore makes it difficult to maintain the classical LES framework in which the geometrical and effective grids are designed to resolved a large percentage of the variance of all dynamically relevant variables with the impacts of RS-SFS interaction on RS evolution well approximated by models that characterize statistically a dominantly forward-cascading interscale dynamics.

V. DISCUSSION AND CONCLUSIONS

The current study has two interrelated motivations: (1) the development of a “discontinuity pollution removal” procedure that removes nonphysical Fourier content arising from boundary discontinuities when the FTF of a bounded analytic signal is taken in a nonperiodic direction, without significantly modifying the signal away from the boundaries (Secs. II and Appendix B), and (2) analysis of the relationship between the same coherent structure in physical space and in Fourier space with data from a reduced physics simulation designed to have key characteristics of flame-turbulence interactions that are characteristic of flame-turbulence interactions in premixed turbulent combustion (Sec. IV). Because discontinuity pollution removal is required preceding our analysis of coherent structure in Fourier space, we use this simulation also to validate the pollution-removal method (Appendix C).

Our discontinuity pollution removal procedure has potential applications that extend well beyond multidimensional Fourier analysis of premixed turbulent combustion DNS, where “discontinuity” is, in reality, a large gradient with magnitude determined by the fixed boundary jump and the resolution of the grid. Based on the mathematical identification of specific Fourier content associated with the nonperiodicity of the signal (Sec. II B), it is both a “pollution removal” and a (partial) signal periodization procedure. “Partial” periodization arises from the tradeoff between (i) the removal of spectral content associated with the nonphysical discontinuity that is created at the boundaries by the bounded Fourier transform of nonperiodic signals, and (ii) the degree of alteration of the original signal away from the boundaries. To fully periodize a signal with our approach requires the subtraction from the original signal of all spectral content of a “discontinuity function” that contains the same boundary discontinuity as in the original signal as well as a linear ramp between the boundaries that impacts the signal everywhere^{23,25}. To avoid this problem, discontinuity removal is confined to high wavenumbers above a user-determined high-pass filter cutoff that localizes the modification of the signal to the boundary discontinuity, thus minimizing changes to the signal away from the boundary. However, application of the high-pass filter leaves behind a residual level of boundary discontinuity with associated residual level of nonperiodicity. We designed a procedure that determines an optimal high-pass filter cutoff as a function of a user-chosen parameter that decides the degree of residual boundary discontinuity (and nonperiodization) vs. the level of modification of the original signal away from the boundaries (Sec. II C). Whereas in Sec. II the discontinuity pollution removal procedure is developed for analytic 1D signals, the approach is extended to 3D signals with one nonperiodic direction in Appendix B. Our approach is useful to other applications where spectral analysis of nonperiodic computational or experimental signals is required, and we expect that our approach can be generalized and improved.

We developed a detailed analysis to study and validate our approach using 2D data from a flame-vortex-train interaction simulation in Appendix C. In the course of the validation analysis, we learn that the optimization process to determine

the filter cutoff wavenumber, together with the choice of the user-defined parameter α , retains large scale unadulterated content while removing the polluting influence of the boundary discontinuity. This process uncovers the true diffusion-induced exponential spectral drop-off at higher wavenumbers that was fully masked by the power-law content of Fourier modes created by the unphysical boundary discontinuity. It now becomes possible to analyze the true multidimensional spectral content of nonperiodic signals. This is necessary for our application to the calculation of the nonlinear triadic (and quadratic) interactions underlying essential nonlinearities, the subject of a future submittal (briefly summarized in Paes *et al.*¹⁰).

Having removed signal pollution in the nonperiodic direction, we analyzed the relationships between coherent structure in physical and Fourier space through the application of systematic localized filtering in Fourier space in context with the multidimensional Fourier representation of scale content (Sec. IV). Collections of Fourier coefficients are shown to be connected to localized coherent structures in physical space, where “coherence” is embedded in the phase correlations that exist among individual Fourier coefficients at different scales in different directions (i.e., positions in Fourier space) underlying the physical-space representation of the coherent structure.

In particular, small-scale coherent structure is organized within a coherent broadband “star-like” pattern in Fourier space that extends from vortex to the flame scales (Sec. IV A). The magnitudes and directional dependences of the Fourier coefficients within the legs of the star are shown to be closely connected with the direction and magnitude of flame-normal gradients of key variables within the corrugated flame front. This observation suggests that Fourier content of flame-turbulence interactions would also be organized in coherent broadband star-like pattern over a wide range of angles due to the wide range of flame corrugations caused by turbulence. The current study shows that, in more complex multiscale turbulence-flame interactions, collections of flame segments with similar flame-normal orientation may be isolated by the systematic application of specific filtering operations in Fourier space developed in the current study. With this understanding, it is possible to analyze DNS studies to relate embedded coherence in multidimensional scale-space to specific nonlinear interactions underlying evolution in physical space¹⁰.

For a consistent interpretation of our reduced-physics simulations in relationship to LES, we defined resolved scales with a 2D Fourier space filter that decomposes scale-based content of fluctuations in key variables into “resolved” vs “sub-filter” scale (RS vs SFS) based on the key criterion underlying LES that the spectral content of the variances of key variables is captured by the effective grid to the 80 – 90% level (Sec. IV B). The LES framework includes the requirement that, statistically, nonlinear interscale interactions transfer information from the larger resolved scales to the smaller, and much less energetic, subfilter scales. In this way, RS evolution is impacted only indirectly by SFS fluctuations. Both of these requirements are necessarily violated in LES of premixed tur-

bulent combustion due to the critical existence of key kinetics that lead to heat release at scales unresolvable by the grid. Consequently, the range of resolved scales is defined based on relative RS variance of momentum and thermal energy fluctuations, but not on species concentrations.

Interestingly, our analysis shows direct correspondence between localized regions of RS Fourier space and coherent structural features in physical space at the resolved scales, implying that it is now possible to isolate the interscale couplings underlying the evolution of specific structural features. Most importantly we have identified the localized regions in 2D Fourier space that correspond to the corrugations in the flame front in the transverse direction in response to forcing by the vortex train, as well as axial gradients associated with the overall planar structure of the flame. In addition, we have shown that the elongated structures within the products that are created as eddies pass through the flame are localized in Fourier space to axial wavenumbers near $k_x = 0$ and spanwise wavenumbers that match the dominant spanwise wavenumbers k_y of the forcing eddies. We anticipate that these general groupings of Fourier modes exist in fully turbulent flame-turbulence interactions with details dependent on the scales and strengths of the turbulence eddies that distort the flame front, making it possible to study nonlinear scale interactions associated with specific features underlying flame-turbulence dynamics in premixed turbulent combustion. The relative variance content of different coherent structures provides the kinematic significance of distinct structural features for the key variables and their relative RS vs SFS content in LES (Sec. IV D). Whereas over 90% of the variance of HCO is subfilter scale, momentum fluctuations are mostly resolved by the effective grid. Although major fluctuations in momentum may be found in the vortices on the reactants side and streaks on the products side of the flame (i.e., the turbulence eddies), significant variance content also exists in the deformations of the flame front (Fig. 24).

In Sec. IV A we explained that two additional computational models have been analyzed with “moderate” and “strong” vortices, where the moderate vortices generated long neck distortions of the flame sheet and strong vortices produce pinching of necks into pockets that continue to burn within the burnt products. As discussed in Paes¹³, there were few findings that add significantly to what has been discussed here. Primarily it was found that the stronger-vortex cases emphasize the kinematic Fourier-physical-space relationships associated with strongly corrugated flame sheets. Specifically, the variance content of flame corrugations (red spectral regions in Fig. 19) increases relative to the vortices. In addition, the star-like pattern becomes buried within frontal contributions induced by discontinuities at the surfaces of the pockets which radiate in all directions and obscure the directions of larger-area flat portions of the primary flame sheet. Extrapolating to fully-developed turbulence, one anticipates that the star-like pattern observed distinctly (Figs. 11 and 12) becomes a roughly isotropic distribution of star-like patterns. In the current study we learn that individual kinematic contributions to Fourier-space structure from frontal content in the presence of the full range of turbulence scales can be extracted with the

application of elliptical filters extended to 3D Fourier space similar to the application of three-dimensional wavelet filtering with physically-modeled wavelet scales (see Brasseur and Wang⁵, for example).

The current analysis provides a deeper understanding of key SFS modeling dynamics in LES of turbulent reacting flows, where RS response to the injection of thermal energy well below grid scale cannot be directly predicted and cannot be modeled assuming classical forward-cascade-dominant dynamics where SFS impacts on RS evolution are largely diffusive. Section IV in the current study provides a deeper understanding of the interscale coupling issues that must be considered in advanced closure strategies. We show, for example, that the SFS variance of all variables analyzed is associated with SFS content of the corrugated flame front. In 2D Fourier space, the flame front lies within the spectral star-like coherent structure, a structure that resides partially in the resolved scales and partially in the subfilter scales. One might therefore anticipate potentially important RS-SFS couplings among the Fourier modes associated with this multiscale coherent structure. We aim to follow the current report with an equally extensive report of the nonlinear couplings associated with the physical-Fourier space coherent structures described in the current study.

ACKNOWLEDGMENTS

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Appendix A: Representation in Physical and Fourier Space of Higher-order Boundary Discontinuities of Nonperiodic Signals in Finite Domains

Consider an arbitrary smooth and continuous 1D signal $f(x)$ over a finite domain $0 \leq x \leq L$ with boundary values $f(0)$ and $f(L)$. As discussed in Sec. II, if $f(L) \neq f(0)$, the periodic extension of $f(x)$ has a C^0 discontinuity at its boundaries with a finite jump $\Delta f = f(L) - f(0)$. As shown in Eq. (3), the C^0 discontinuity is characterized by a linear “discontinuity function” $d(x)$ with Fourier coefficients $\hat{d}(k_x)$ that have power-law

dependence as k_x (Eq. (2)). This description may be generalized to higher order boundary discontinuities C^N with jumps in the n^{th} derivative $f^{(n)}(x) \equiv d^n f/dx^n$ at the boundary given by $\Delta f^{(n)} = f^{(n)}(L) - f^{(n)}(0)$. These higher order discontinuities are represented in physical space by linear discontinuity functions at the (n) derivative level of $f(x)$ which, when integrated back to (0) derivative level of $f(x)$, result in n^{th} order polynomials.

Similarly to the C^0 discontinuity function given by Eqs. 3 and 2, we define the C^N discontinuity function at the derivative level (n) as $d_N^{(n)}(x)$. By construction, the C^N discontinuity function at its highest derivative level $(n = N)$ contains the same discontinuity as $f^{(N)}(x)$ separated by a linear function as per Eq. (3) at the functional level:

$$d_N^{(N)}(x) = \frac{\Delta f^{(N)}}{L}x + \mathbb{C}_N^{(N)}. \quad (\text{A1})$$

To construct the C^N discontinuity function at the function level $d_N^{(0)}(x)$ ($n = 0$), $d_N^{(N)}(x)$ is integrated sequentially from $d_N^{(N)}(x)$ to $d_N^{(N-1)}(x)$, then to $d_N^{(N-2)}(x)$, $d_N^{(N-3)}(x)$, ..., $d_N^{(n)}(x)$, ..., ending at the function level $d_N^{(0)}(x)$. At each integration level a new constant of integration is introduced. These are found with three requirements. First, the discontinuity in the integrated discontinuity function at level $d_N^{(n)}(x)$ must equal the discontinuity in the n^{th} derivative in the function $f(x)$ at the n^{th} level:

$$\Delta d_N^{(n)} \equiv d_N^{(n)}(L) - d_N^{(n)}(0) = \Delta f^{(n)} \quad (\text{A2})$$

Second, at the functional ($n = 0$) level, all discontinuity functions must have the same boundary values, so that at $x = 0$, $d_N^{(0)}(0) = d_{N-1}^{(0)}(0) = d_{N-2}^{(0)}(0) = d_0^{(0)}(0)$. Finally, as discussed in context with Eq. (2), the discontinuity function at the functional level has zero mean so that $\widehat{d_N^{(0)}}(k_x = 0) = 0$. The result is a collection of polynomial functions $d_N^{(0)}(x)$ that contain $C^{M \leq N}$ discontinuities at the boundaries, $M = 0, \dots, N$.

Using the approach just described, the C^0 discontinuity is represented by a linear function at its $n = N = 0$ derivative level,

$$d_0^{(0)}(x) = \frac{\Delta f^{(0)}}{L}x + \mathbb{C}_0^{(0)} = \frac{\Delta f^{(0)}}{L}(x - \frac{L}{2}), \quad (\text{A3})$$

where the constant $\mathbb{C}_0^{(0)} = -\frac{\Delta f^{(0)}}{2}$ is so that $d_0^{(0)}(x)$ has zero mean (Section II A).

Similarly, the C^1 discontinuity is represented by a linear function at the $n = N = 1$ derivative level:

$$d_1^{(1)}(x) = \frac{\Delta f^{(1)}}{L}x + \mathbb{C}_1^{(1)}, \quad (\text{A4})$$

which, when integrated results in

$$d_1^{(0)}(x) = \int d_1^{(1)}dx + \mathbb{C}_1^{(0)} = \frac{\Delta f^{(1)}}{L} \frac{x^2}{2} + \mathbb{C}_1^{(1)}x + \mathbb{C}_1^{(0)}, \quad (\text{A5})$$

where $\mathbb{C}_1^{(0)} = \mathbb{C}_0^{(0)} = -\frac{\Delta f^{(0)}}{2}$ is obtained by requiring that $d_1^{(0)}(0) = d_0^{(0)}(0)$. The constant $\mathbb{C}_1^{(1)}$ is found by requiring that $\Delta d_1^{(0)} = \Delta f^{(1)}$ in Eq. (A5), so that $\mathbb{C}_1^{(1)} = \frac{\Delta f^{(0)}}{L} - \frac{\Delta f^{(1)}}{2}$. Thus, the C^0 and C^1 boundary discontinuities in $f(x)$ are represented by a combination of parabolic and linear functions:

$$d_1^{(0)}(x) = \frac{\Delta f^{(1)}}{L}x \left(\frac{x-L}{2} \right) + d_0^{(0)}(x). \quad (\text{A6})$$

We now represent the C^2 discontinuity by a linear function at the $n = N = 2$ derivative level:

$$d_2^{(2)}(x) = \frac{\Delta f^{(2)}}{L}x + \mathbb{C}_2^{(2)}. \quad (\text{A7})$$

When integrated twice this produces

$$d_2^{(1)}(x) = \int d_2^{(2)}dx + \mathbb{C}_2^{(1)} = \frac{\Delta f^{(2)}}{L} \frac{x^2}{2} + \mathbb{C}_2^{(2)}x + \mathbb{C}_2^{(1)}, \quad (\text{A8})$$

and

$$d_2^{(0)}(x) = \int d_2^{(1)}dx + \mathbb{C}_2^{(0)} = \frac{\Delta f^{(2)}}{L} \frac{x^3}{6} + \mathbb{C}_2^{(2)} \frac{x^2}{2} + \mathbb{C}_2^{(1)}x + \mathbb{C}_2^{(0)}. \quad (\text{A9})$$

where $\mathbb{C}_2^{(0)} = \mathbb{C}_1^{(0)} = \mathbb{C}_0^{(0)} = -\frac{\Delta f^{(0)}}{2}$ by setting $d_2^{(0)}(0) = d_1^{(0)}(0) = d_0^{(0)}(0)$.

The constant $\mathbb{C}_2^{(2)}$ is found by requiring that $\Delta d_2^{(1)} = \Delta f^{(1)}$ in Eq. (A8), so that $\mathbb{C}_2^{(2)} = \frac{\Delta f^{(1)}}{L} - \frac{\Delta f^{(2)}}{2}$. Similarly, $\mathbb{C}_2^{(1)}$ is found by requiring that $\Delta d_2^{(0)} = \Delta f^{(0)}$ in Eq. (A9), so that $\mathbb{C}_2^{(1)} = \frac{\Delta f^{(0)}}{L} - \frac{\Delta f^{(1)}}{2} + \Delta f^{(2)} \frac{L}{12}$. Thus, the C^0 , C^1 , and C^2 boundary discontinuities in $f(x)$ are represented by a combination of polynomial discontinuity functions:

$$d_2^{(0)}(x) = \frac{\Delta f^{(2)}}{L}x \left(\frac{x^2}{6} - \frac{xL}{4} + \frac{L^2}{12} \right) + d_1^{(0)}(x) \quad (\text{A10})$$

The procedure to represent the fundamental discontinuities at the boundaries in physical space may be extended to arbitrary order discontinuities $C^{1 \leq M \leq N}$ as previously discussed. First, the C^N discontinuity is represented by a linear function $d_N^{(N)}(x)$ at the $n = N$ derivative level as per Eq. (A1), which introduces an arbitrary constant $\mathbb{C}_N^{(N)}$. This linear function is incrementally integrated N times to recover the polynomial $d_N^{(0)}(x)$ at the (0) derivative level, which introduces N arbitrary constants $\mathbb{C}_N^{(M)}$ associated with the integrated functions $d_N^{(M)}$ for $M = N-1, \dots, 0$, respectively. The constant $\mathbb{C}_N^{(0)} = \mathbb{C}_0^{(0)} = -\frac{\Delta f^{(0)}}{2}$ is obtained by requiring that $d_N^{(0)}(0) = d_0^{(0)}(0)$. Starting at the highest M level of $\Delta d_N^{(M)}$, each unknown constant $\mathbb{C}_N^{(M)}$ is found by requiring that $\Delta d_N^{(M)} = \Delta f^{(M)}$ in the respective integrated level of $d_N^{(N)}(x)$ as per Eq. (A2).

In Fourier space, Eq. (1) may be generalized to $f^{(n)}(x)$,

$$\begin{aligned} \widehat{f^{(n)}}(k_x \neq 0) &= \frac{1}{L} \int_0^L f^{(n)}(x) e^{-ik_x x} dx \\ &= -\frac{i}{k_x} \widehat{f^{(n+1)}}(k_x) + \frac{i}{k_x} \frac{\Delta f^{(n)}}{L}. \end{aligned} \quad (\text{A11})$$

The fundamental $C^{M \leq N}$ discontinuities at the boundaries are therefore represented in Fourier space as

$$\widehat{d}_N^{(0)}(k_x \neq 0) = \sum_{n=0}^N \frac{\Delta f^{(n)}}{L} \left(\frac{i}{k_x} \right)^{n+1}, \quad N \geq 0, \quad (\text{A12})$$

where $\widehat{d}_N^{(0)}(0) = 0$.

Appendix B: Extension of “Discontinuity Pollution Removal” Procedure to Multi-Dimensional Signal

1. Basic Strategy

To apply the multidimensional Fourier description to the nonperiodic signals described in Sec. III C, the nonphysical spectral content that arises from discontinuities at the boundaries in x must be removed using the “discontinuity pollution removal” procedure described in Sec. II C. Here we extend the procedure to multi-dimensional signals with only one non-periodic direction. Whereas we develop the generalization to 3D signals, the application to 2D is straightforward.

Consider an analytic 3D signal $f(x, y, z)$ bounded between $(0, 0, 0)$ and (L_x, L_y, L_z) . The signal is periodic in y and z and nonperiodic in x , so that the boundary discontinuity $\Delta f(y, z) = f(L_x, y, z) - f(0, y, z)$ can vary with (y, z) . There are two ways that the discontinuity removal process in x described in Sec. II C might be applied. In the first, the 1D procedure developed there is applied independently along x pencils at discretized (y, z) locations on the transverse plane. This, however, leads to discontinuities in the modified signal over the (y, z) plane that we would like to avoid. We therefore apply the second method, whereby the 1D procedure described in Sec. II C is developed for the two-dimensional bounded FTF of $f(x, y, z)$ in y and z :

$$\widehat{f}^{y,z}(x; k_y, k_z) \equiv FTF_{y,z}\{f(x, y, z)\}, \quad (\text{B1})$$

where we use the hat with adjacent coordinates to indicate the taking of a Fourier transform in (y, z) coordinate directions. The Fourier coefficients are complex functions defined at discrete transverse wavevectors with discrete components (k_y, k_z) that contain the boundary jumps in x reflecting nonperiodicity as a function of (k_y, k_z) . In this way, the discontinuity removal process is decomposed in terms of spatial scale globally over the transverse plane.

Following the procedure in Sec. II A, integration by parts of the Fourier transform of Eq. (B1) in the nonperiodic direction x is

$$\begin{aligned} \widehat{f}^{y,z}(k_x; k_y, k_z) &= \frac{1}{L_x} \int_0^{L_x} \widehat{f}^{y,z}(x; k_y, k_z) e^{-ik_x x} dx \\ &= -\frac{i}{k_x} \frac{\partial \widehat{f}^{y,z}}{\partial x}(k_x; k_y, k_z) + \frac{i}{k_x} \frac{\Delta \widehat{f}^{y,z}(k_y, k_z)}{L_x} \end{aligned} \quad (\text{B2})$$

for $k_x \neq 0$, where we use the second hat with adjacent coordinate x to indicate the Fourier transform is in x . The “discontinuity function” is therefore

$$\widehat{d}^{y,z}(k_x; k_y, k_z) = \frac{i}{k_x} \frac{\Delta \widehat{f}^{y,z}(k_y, k_z)}{L_x}, \text{ for } k_x \neq 0, \quad (\text{B3})$$

with $\widehat{d}^{y,z}(0; k_y, k_z) = 0$ (zero mean in x). This has the following inverse transform in x :

$$\widehat{d}^{y,z}(x; k_y, k_z) = \frac{\Delta \widehat{f}^{y,z}(k_y, k_z)}{L_x} \left(x - \frac{L}{2} \right). \quad (\text{B4})$$

Thus the discontinuity function in Fourier space, $\widehat{d}^{y,z}(k_x; k_y, k_z)$, is represented in physical space by a sawtooth function with zero mean. However, unlike the treatment for real 1D functions in Sec. II, here the discontinuity function is complex similar to the 2D Fourier transform of $f(x, y, z)$.

Following the developments in Sec. II C, to localize near the discontinuities at the boundaries $\widehat{d}^{y,z}(x; k_y, k_z)$ is high-pass filtered to produce a modified discontinuity function with suppressed low-wavenumber contributions from the linear ramp in the discontinuity function between the two boundaries:

$$\widehat{d}_m^{y,z}(k_x; k_y, k_z, k_{cut}) = m(k_x; k_{cut}) \widehat{d}^{y,z}(k_x; k_y, k_z), \quad (\text{B5})$$

where $m(k_x; k_{cut})$ is the high-pass filter, equal to one when $k_x \geq k_{cut}$ and zero when $k_x < k_{cut}$. Note that $k_{cut} = k_{cut}(k_y, k_z)$, so k_{cut} is determined for each discrete wavevector (k_y, k_z) on the plane perpendicular to the nonperiodic direction. The inverse transform in x of Eq. (B5) returns the function to physical space variations in x :

$$\widehat{d}_m^{y,z}(x; k_y, k_z, k_{cut}) = FTF_x^{-1} \left\{ \widehat{d}_m^{y,z}(k_x; k_y, k_z, k_{cut}) \right\}. \quad (\text{B6})$$

$\widehat{d}_m^{y,z}(x; k_y, k_z, k_{cut})$ is a modified version of the discontinuity function that localizes to the discontinuities and suppresses the linear ramp of the sawtooth.

Once a useful optimization algorithm is developed to determine $k_{cut}(k_y, k_z)$ (below), the FTF of the original signal $f(x, y, z)$ is modified to remove spectral content arising from the x boundary discontinuities within f :

$$\widehat{f}_m^{y,z}(k_x; k_y, k_z, k_{cut}) = \widehat{f}^{y,z}(k_x; k_y, k_z) - \widehat{d}_m^{y,z}(k_x; k_y, k_z, k_{cut}). \quad (\text{B7})$$

In physical space the modified signal has the form

$$f_m(x, y, z, k_{cut}) = f(x, y, z) - d_m(x, y, z, k_{cut}), \quad (\text{B8})$$

where each term in Eq. (B8) is the 3D inverse Fourier transform of the corresponding Fourier coefficients in Eq. (B7). The residual boundary jump in x in the modified signal is therefore given by

$$\begin{aligned} \Delta f_m(y, z, k_{cut}) &= f_m(L_x, y, z, k_{cut}) - f_m(0, y, z, k_{cut}) \\ &= \Delta f(y, z) - \Delta d_m(y, z, k_{cut}), \end{aligned} \quad (\text{B9})$$

where $\Delta f(y, z)$ and $\Delta d_m(y, z, k_{cut})$ are the boundary jumps in the original signal and modified discontinuity function, respectively. Ultimately, we analyze $\widehat{f}_m^{y,z}(k_x; k_y, k_z, k_{cut})$ in Fourier space and $f_m(x, y, z, k_{cut})$ in physical space.

2. Algorithm to Determine the Filter Cutoffs

To complete the discontinuity removal process, it is necessary to develop an algorithm to determine the filter cutoff $k_{cut}(k_y, k_z)$ for each discrete wavevector (k_y, k_z) . In doing so, one recognizes that, because all functions are real in physical space, the reality condition, $\hat{h}(-k_y^+, -k_z^+) = \hat{h}^*(k_y^+, k_z^+)$ must be maintained for all functions $h(y, z)$. Thus, for every “positive” wavevector (k_y^+, k_z^+) there exists a “negative” wavevector $(-k_y^+, -k_z^+)$ with a Fourier coefficient determined by the Fourier coefficient of the “positive” wavevector (k_y^+, k_z^+) and $k_{cut}(-k_y^+, -k_z^+) = k_{cut}(k_y^+, k_z^+)$. How one separates the set of wavevectors (k_y, k_z) into the sets (k_y^+, k_z^+) and $(-k_y^+, -k_z^+)$ is, in principle, arbitrary. The algorithm to determine $k_{cut}(k_y, k_z)$ for each (k_y^+, k_z^+) pencil is developed in physical space by extracting the Fourier coefficients in k_x of $\widehat{d_m^{y,z^x}}(k_x; k_y, k_z, k_{cut})$ and $\widehat{f_m^{y,z^x}}(k_x; k_y, k_z, k_{cut})$ for each pair of pencils emanating in x from the positive and negative wavevectors, (k_y^+, k_z^+) and $(-k_y^+, -k_z^+)$:

$$\tilde{h}(x; y, z) = FTF_{x,y,z}^{-1} \left\{ \delta(|k_y| - k_y^+, |k_z| - k_z^+) \widehat{h^{y,z^x}}(k_x; k_y, k_z) \right\}. \quad (\text{B10})$$

The tilde ($\sim$) implies that the dependence of $\tilde{h}(x; y, z)$ on y and z comes from a single harmonic Fourier mode selected by the Dirac delta function that defines a sine wave on the $y-z$ plane with wavelength and direction determined by the magnitude and direction of positive/negative wavevector pair associated with the Fourier mode. The x -dependence, on the other hand, comes from the x -dependence in the nonperiodic direction of the original signal and in $\widehat{h^{y,z^x}}(k_x; k_y, k_z) \sim \widehat{f^{y,z^x}}$, $\widehat{d_m^{y,z^x}}$, and $\widehat{f_m^{y,z^x}}$ in Eq. (B10), including the residual boundary jump in $\widehat{f_m^{y,z^x}}$ along scale-based pencils in x at (k_y^+, k_z^+) and $(-k_y^+, -k_z^+)$.

The aim is to minimize the residual boundary jump in f_m (Eq. (B9)) without significantly altering the signal away from the x boundaries. To this end, we follow the approach described in Sec. II C whereby a weighting parameter α (Eq. (10)) is used to balance two parameters, one designed to quantify the relative change made in the signal away from the boundary (ϵ , Eq. (7)) and the other designed to quantify the level of residual boundary discontinuity and non-periodicity in x (Δ_r , Eq. (8)) retained in the modified signal, f_m . In the multidimensional application, however, these parameters are formed using the form of the function $\tilde{h}$ in Eq. (B10) that select a single pair of Fourier modes on the $y-z$ plane defined by the “positive” mode, (k_y^+, k_z^+) :

$$\epsilon(k_y^+, k_z^+; k_{cut}) = \frac{\|\widehat{d_m^{y,z^x}}(x, y, z; k_{cut})\|_V}{\|\tilde{f}(x, y, z)\|_V}, \quad (\text{B11})$$

$$\Delta_r(k_y^+, k_z^+; k_{cut}) = \max_{y,z} \left\{ \frac{|\Delta \widehat{f_m^{y,z^x}}(y, z; k_{cut})|}{\|\max_x (\partial \tilde{f}(x, y, z) / \partial x)\| \Delta x} \right\} \quad (\text{B12})$$

where $k_{cut} = k_{cut}(k_y^+, k_z^+)$. Both parameters are defined for pencils at (k_y^+, k_z^+) with their conjugate locations $(-k_y^+, -k_z^+)$. In Eq. (B11), $\|\dots\|_V$ is the L_2 norm over volume V within the computational domain, allowing the user to select, if desired, the region that is most critical to minimize deviation in the original signal. In Eq. (B12), $\Delta \widehat{f_m^{y,z^x}}(y, z, k_{cut}) = \widehat{f_m^{y,z^x}}(L_x, y, z, k_{cut}) - \widehat{f_m^{y,z^x}}(0, y, z, k_{cut})$, defined by inserting $\widehat{f^{y,z^x}}$ into Eq. (B9) for $\widehat{h^{y,z^x}}(k_x; k_y, k_z)$, is a function of (y, z) . The denominator selects the largest physical x gradient in $\tilde{f}$ in x away from the boundaries, where $\tilde{f}$ is defined along (k_y, k_z) pencils. Both numerator and denominator of the ratio in Eq. (B12) depend on (y, z) and Δ_r is defined as the maximum value of the ratio in Eq. (B12) over the $y-z$ plane.

$\epsilon(k_y^+, k_z^+; k_{cut})$ and $\Delta_r(k_y^+, k_z^+; k_{cut})$ are both functions of k_{cut} , which is itself a function of (k_y^+, k_z^+) . As per Sec. II C, we select the optimal value for k_{cut} at each (k_y^+, k_z^+) by identifying the minimum in the following function with a specified weighting parameter, α :

$$\phi(k_{cut}; \alpha) = \alpha \epsilon(k_y^+, k_z^+; k_{cut}) + (1 - \alpha) \Delta_r(k_y^+, k_z^+; k_{cut}). \quad (\text{B13})$$

The optimal choice for k_{cut} is found at the minimum in ϕ as a function of k_{cut} for specified fixed α :

$$\phi(k_{cut}^{opt}; \alpha) = \min_{k_{cut}} [\phi(k_{cut}; \alpha)]. \quad (\text{B14})$$

As described in Sec. II C, α is chosen by the user to balance the degree of reduction of the polluting consequences of the unphysical boundary discontinuity induced by the Fourier transform in the nonperiodic direction with the level of modification to the signal away from the boundary.

The result of applying Eq. (B14) with user-defined α is an optimal value of k_{cut} for each value of (k_y, k_z) as needed to complete the discontinuity removal process extended to multi-dimensional signals as described above in Appendix B 1.

Appendix C: Validation and Parameterization of the Discontinuity Pollution Removal Procedure

The discontinuity removal procedure introduced in Sec. II and formalized for multi-dimensional signals in Appendix B is validated using the 2D flame-vortex interaction simulation described in Sec. III C. To illustrate the outcome of the procedure, Fig. 25 presents the original (f) and modified ($f_m = f - d_m$) signals of ρu along with its associated modulated discontinuity function, d_m , in physical space. The signals are shown over the computational domain ($0 \leq x/D \leq 16$) at the time instant of largest boundary discontinuity in x (Δf) in ρu , which occurs with the vortices at the inflow and elongated streaks in the products (Sec. III C). Note that in Secs. III C and IV, the signals are analyzed at the time instant when the flame is most “corrugated”.

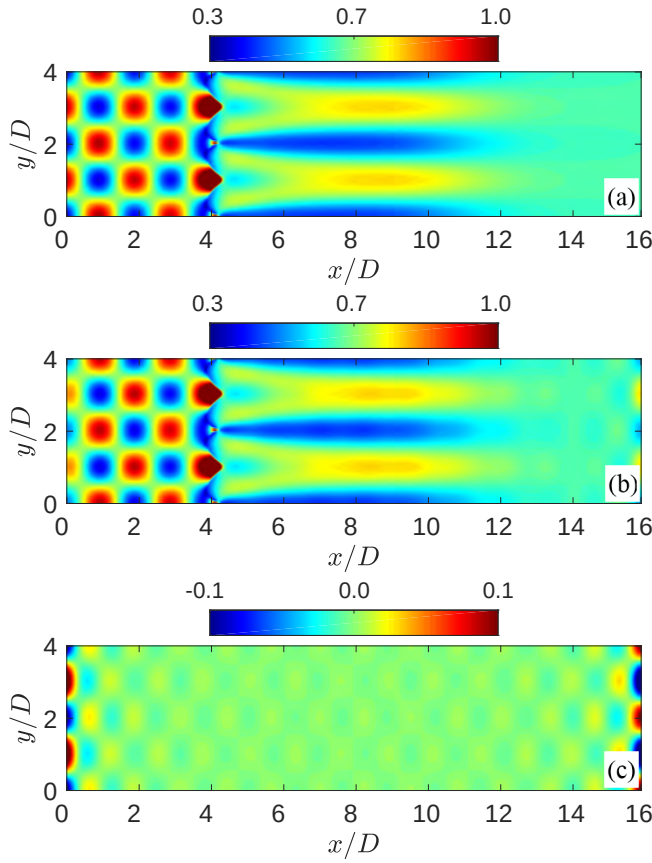


FIG. 25: Isocontours of (a) original signal $f(x,y)$ and (b) modified signal $f_m(x,y)$ in physical space of ρu for the 2D flame-vortex interaction simulation. The associated modulated discontinuity function d_m is shown in (c).

The Fourier-space counterparts of the original, post-processed, and modulated discontinuity signals associated with ρu are presented in Fig. 26. We plot isocontours of the logarithms of squared Fourier coefficients of ρu with the color bars spanning 5 orders of magnitude of $\log(\rho u)^2$. The wavenumber axes are normalized using the planar laminar flame thickness. The figure shows 2D Fourier space down

to the flame scale, which are the values $k_{x,y}\delta_f/\pi = \pm 1$. The pink box identifies the vortex scale defined by $k_{x,y}\delta_f/\pi = \pm \delta_f/D = 0.06$, where $k_v \equiv \pi/D$ is the vortex-scale wavenumber (see Fig. 9b), the four Fourier modes used to construct the train of vortices at inflow. As explained in Sec. IV, the spanwise symmetry of the inflow vortices leads to negligible Fourier content in Fourier modes odd in k_y (i.e., at $k_y/\Delta k_y = \pm 1, \pm 3, \dots$). These odd k_y modes have been removed from all spectral isocontour plots to improve the visualization.

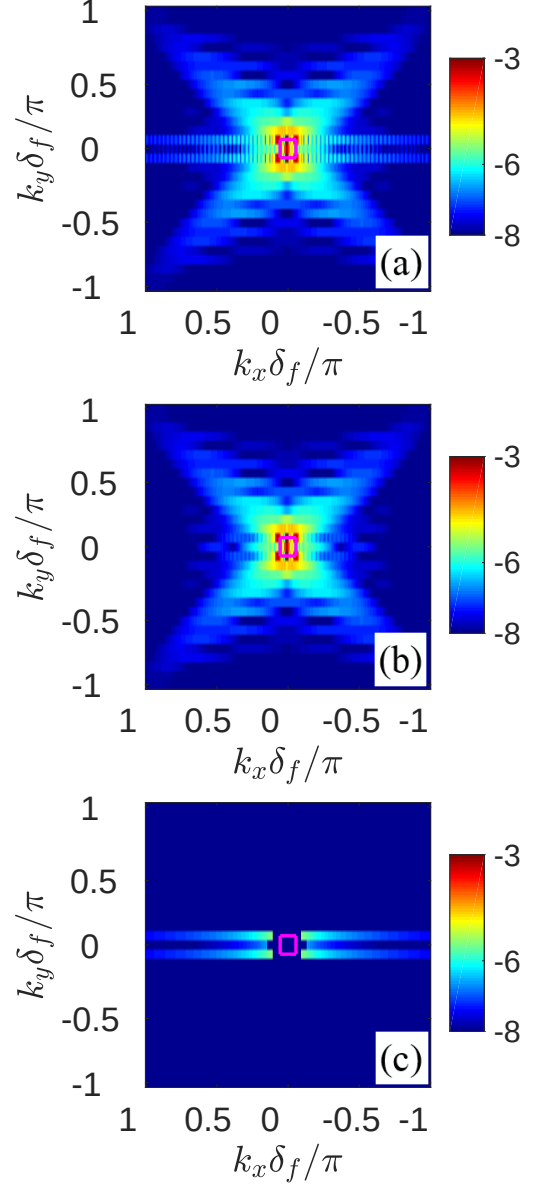


FIG. 26: Isocontours of the (a) original $\hat{f}(k_x; k_y, k_z)$ and (b) modified $\hat{f}_m(k_x; k_y, k_z, k_{cut})$ signal in Fourier space of $\log(\rho u^2)$ for the 2D flame-vortex interaction simulation. The associated modulated discontinuity is shown in (c).

The highest magnitude Fourier energy content in Fig. 26a includes a “star-like” pattern that extends to the highest wavenumbers, extending from the vortex scale box to the

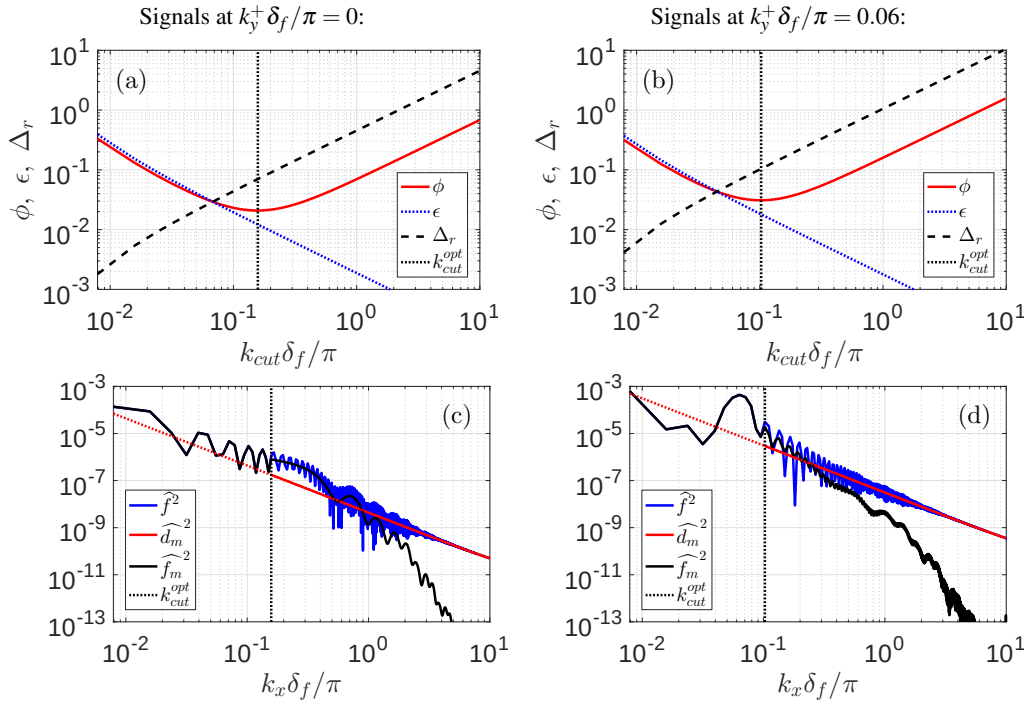


FIG. 27: (a-b) Computed ϕ , ϵ , and Δ_r as a function of k_{cut} for $\alpha = 0.85$ and V in the range $2 \leq x/D \leq 8$; (c-d) Squared Fourier coefficients $\hat{f}^2$, $\hat{d}_m^2$, and $\hat{f}_m^2$ as a function of k_x , where dashed red line indicates unmodulated $\hat{d}^2$. Signals at $k_y^+ \delta_f / \pi = 0$ (left: a, c) and 0.06 (right: b, d). The vertical dotted line indicates optimal k_{cut} .

flame scale box aligned at $\pm 42^\circ$ from the k_x -axis. In Sec. II A we showed that discontinuities create power-laws in Fourier space that extend from the lowest wavenumbers to the grid scale, where a discretized discontinuity becomes a large gradient. The star-like pattern in Fig. 26 arises from power-law drops in variance arising from the large physical gradients at the flame front, as described next in Sec. IV. While the star-like pattern is associated with physically-relevant frontal structures of the signals (i.e., the corrugated flame fronts in Fig. 10a), unphysical power-law variations associated with boundary discontinuities are observed in Fig. 26b as bands over k_x with large Fourier content at $|k_y| \delta_f / \pi \leq 0.06$ (i.e., at wavenumbers $k_y / \Delta k_y = 0, \pm 2$). These scales are associated with the cross-stream averaged mean ($k_y \delta_f / \pi = 0$) and the transverse scale of the vortices ($k_y \delta_f / \pi = \pm \delta_f / D$), which introduce boundary discontinuities between inflow and outflow (Fig. 25a).

To effectively remove this pollution, the discontinuity removal procedure described in Appendix B was applied with $\alpha = 0.85$, which ensures that the maximum signal modification at the polluted wavenumbers (Eq. (B11)) remains smaller than 5%. The fundamental discontinuity function $d(x, y)$ is constructed considering only the C^0 discontinuity (Eq. (B3)) and the volume of interest in Eqs. B11 and B12 is the region $2 \leq x/D \leq 8$.

Figures 25b-c and 26b-c show in physical and Fourier space, respectively, the 2D instantaneous isocontours of the modulated discontinuity function (d_m) and the post-processed signal ($f_m = f - d_m$). The modulated discontinuity function

in physical space (Fig. 25c) minimizes the modification in the signal within the domain and localizes the harmonic functions towards the discontinuities at the boundaries in x with variations at the vortex scale in y . Thus, the final signal in physical space (Fig. 25b) preserves the original signal (Fig. 25a) away from the boundaries. In Fourier space, the modulated discontinuity function (Fig. 26c) captures the power-law pollution in k_x at $k_y \delta_f / \pi = 0$ and ± 0.06 with negligible contribution at unpolluted wavenumbers. The optimal cutoff wavenumbers $k_{cut}^{opt}(k_y^+)$ that modulate the power-laws in Fig. 26c is higher than the k_x vortex-scale wavenumber, so that the power-law pollution is removed at scales smaller than the vortex scales ($|k_x| \delta_f / \pi > 0.06$). Consequently, while power-law pollution from the boundary discontinuity is removed in the modulated signal in Figs. 26b and 25b, the large-scale vortex structures and the star-like pattern associated with the corrugated flame front are unaffected.

To access the tradeoff between the signal modification and the residual boundary discontinuity in the selection of the optimal wavenumber cutoffs $k_{cut}^{opt}(k_y^+)$, we analyze the filtered signals at each k_y^+ separately (Eq. (B10)). Figures 27a-b show at fixed $k_y^+ \delta_f / \pi = 0$ and 0.06, respectively, the computed ϕ , ϵ , and Δ_r as a function of k_{cut} , where the optimal wavenumber cutoff $k_{cut}^{opt}(k_y^+)$ is indicated by the vertical dotted black lines. Figure 27c-d and 28a-b show, in Fourier and physical space ($y/D = 1$) respectively, the filtered signals associated with f , d_m , and f_m for $k_y^+ \delta_f / \pi = 0$ and 0.06.

In Fourier space, the original signal shows a clear power-law pollution at the highest wavenumbers starting at $k_x \delta_f / \pi \approx$

1 for $k_y^+ \delta_f / \pi = 0$ (Fig. 27c) and at $k_x \delta_f / \pi \approx 0.2$ for $k_y^+ \delta_f / \pi = 0.06$ (Fig. 27d). Above these polluted wavenumbers Δ_r crosses 1 in Fig. 27a-b and Fourier coefficients are modified by the procedure without significant modification of the original signal, as indicated by the values of $\varepsilon < 2\%$. (Note that ε is as high as 40% at the smallest k_{cut} .) The optimal wavenumber cutoffs k_{cut}^{opt} with $\alpha = 0.85$ (vertical black dotted lines in Fig. 27) favors the minimization of signal modification, so that $\varepsilon(k_{cut}^{opt}) \sim 1-2\%$ and $\Delta_r(k_{cut}^{opt}) \sim 9-15\%$ for $k_y^+ \delta_f / \pi = 0, 0.06$. As a result, the Fourier energy of the modified signal in Fig. 27c-d transitions to exponential decay around the flame front thickness ($k_x \delta_f / \pi \approx 1$) with approximate power decay exponents of -8.60 and -7.52, respectively. Thus, the discontinuity removal process has recovered the signal associated with the flame front. As importantly, the Fourier content associated with the vortex scales (the peak in the spectrum of f in Fig. 27d at $k_y^+ \delta_f / \pi = 0.06$) is preserved.

In Fig. 28 we plot two examples of physical space functions $\tilde{f}(x, y)$, $\tilde{d}_m(x, y)$, and $\tilde{f}_m(x, y)$, filtered as per Eq. (B10) for pencils in x at $k_y^+ \delta_f / \pi = 0$ and 0.06, shown as a function of x/D at $y/D = 1$. Variations in x/D at plotted at $y/D = 1$. The modified signal f_m is shown to preserve the original signal away from the boundaries with modification towards the domain boundaries ($\tilde{d}_m(x; k_y^+)$ in Fig. 28). The “ripples” result from the sharp filter $m(k_x; k_{cut})$ (Eq. (6)) employed to modulate $\tilde{d}(k_x)$. This kinematic ringing effect can be removed with the use of smooth Fourier space filters (not shown). Note, from Fig. 28 that the vortices in the reactants ($x/D < 4$), elongated streaks in the products ($x/D > 4$), and large gradients within the flame front ($x/D \approx 4$) are all preserved in the modified signal.

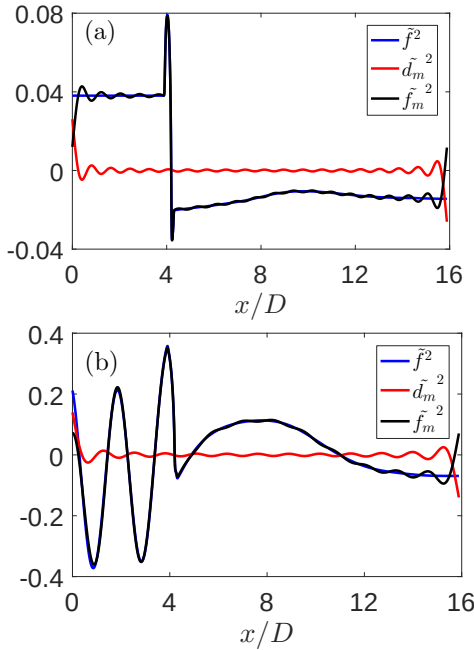


FIG. 28: $\tilde{f}$, $\tilde{d}_m$, and $\tilde{f}_m$, filtered as per Eq. (B10) at (a) $k_y^+ \delta_f / \pi = 0$ and (b) 0.06, shown as a function of x/D at $y/D = 1$.

Figure 29 shows the sensitivity of the maximum $\varepsilon(k_y^+)$ (solid curves) and $\Delta_r(k_y^+)$ (dashed curves) as a function of α for the pollution removal procedure applied to ρu , ρv , u , and ρh from the 2D flame-vortex interaction simulation. Because ρY_{HCO} is highly localized within the flame $\Delta f = 0$. Therefore $d = 0$ is zero and the signal is unaffected. The procedure is shown to be robust for distinct variables and the choice of $\alpha = 0.85$ ensures that the maximum signal modification and residual jump in the post-processed signal for all variables remains smaller than 5% and 30%, respectively, as indicated by the horizontal lines in Fig. 29.

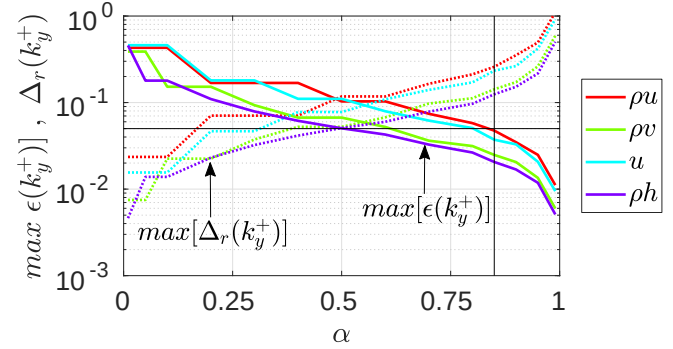


FIG. 29: Maximum over k_y^+ of ε (solid curves) and Δ_r (dashed curves) as a function of α for different variables in the 2D flame-vortex interaction simulation. The vertical lines is at $\alpha = 0.85$.

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Appendix B

A Novel Strategy to Identify Dynamically Dominant Inter-Scale Couplings for Application to Large-Eddy Simulation of Premixed Turbulent Combustion

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Large-Eddy Simulation (LES) is a powerful formulation for model turbulent reacting flows that balances lower resolution with predictions of variance dominant momentum and energy fluctuations. Central to the LES framework is the need to model nonlinear interactions between the variance-dominant resolved-scale (RS) motions and subfilter-scale (SFS) fluctuations. A central feature of premixed turbulent combustion is nonlinear flame-turbulence interactions, a key element that involves advective nonlinear coupling between velocity and momentum, energy and species concentration. In this work, we extract the dominant RS-SFS advective nonlinear interactions to isolate SFS content that underlies the evolution of the resolved scales for encapsulation within mathematical forms for potential use in advanced modeling strategies. We take advantage of the mathematical simplicity of the Fourier spectral description of the equations of motion where advective nonlinearities are given by a sum of “triadic” interactions each of which involves three Fourier modes whose wave-vectors form a triangle in multidimensional Fourier space. This elemental representation of key nonlinearities is used to develop a novel strategy to arrange and down-select the dominant nonlinear RS-SFS couplings underlying the evolution of RS motions, and extract the corresponding SFS content associated with dynamically dominant RS-SFS dynamics. The procedure is applied to analysis of “reduced physics” simulations of flame-vortex interactions with a new method for analysis of non-periodic signals in multi-dimensional Fourier-space. For primary variables with dominant variance in the grid-resolved scales and fast decay of energy into the subfilter scales, the resolved-scale features of the dynamically dominant SFS content are shown to be coupled with the smaller resolved scales characterizing the corrugations and thickness of the RS flame front. In contrast, the dynamically dominant SFS content of intermediate species involved in heat release extend farther in Fourier space to smaller scales. These SFS fluctuations are shown to follow the smallest flame-front corrugations, which deviate from the RS flame geometry in regions with high curvature, such as the flame cusps. The results of this study provide insights relevant to advanced modeling strategies designed to capture key RS-SFS couplings into prediction of RS dynamics.

I. The Large-Eddy Simulation Application in Premixed Turbulent Combustion

Computational Fluid Dynamics (CFD) has been used as an important tool for the design of next generation advanced combustion devices for aerospace applications [1]. With ever-increasing power and advances in computing technologies, Large-Eddy Simulations (LES) is becoming a feasible advanced tool for use in the design of practical aerospace systems and devices [2–5]. In the classical LES framework, the numerical solution of the dynamical equations must be carried out on a grid that resolves well a large percentage of the variances of the most energetic scales of motion under the assumption that those turbulence variables are dominated by the forward cascade of momentum and energy from larger to smaller turbulence length scales [6]. If this is the case, the evolution of the more energetic grid-filtered motions are dominated by the dynamical interactions explicitly resolved on the grid, and the dynamical effects of the unresolved

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Sub-Filter-Scale (SFS) motions on the evolution of the Resolved-Scale (RS) motions are higher order. When this is the case, first and second order statistics are accurately predicted in LES. (In contrast, Reynolds-averaged Navier-Stokes simulations are characterized by the need to model second order statistics at the first order level.)

However, the predictive capabilities of LES for premixed turbulent combustion systems are limited by fundamental deviations from the classical LES framework. In these systems, the chemical reactions dominantly associated with the chemical generation of thermal energy (i.e., heat release), generally occur at scales unresolvable by the LES grid [7, 8]. Consequently, some key dynamics underlying the function of combustion devices are almost always dominantly in the unresolved SFS motions so that basic assumptions that make LES an accurate predictive tool break down. Furthermore, the topological structure of the flame is necessarily frontal in nature, which poses difficulties for an LES strategy that must model coherent structures that live partially in the resolved and subfilter scales with a method that treats turbulence eddies as resolved or subfilter scale motions.

Despite the advancements in combustion modeling [9], most of the existent closure models for turbulent reacting flows rely on some assumptions about the turbulence dynamics and the scale separation between different combustion and turbulence processes. Some modeling procedures consider even more severe simplifications since they were developed for non-reacting, constant density flows [10–12]. As a result, these models have demonstrated inconsistencies when applied to practical combustion problems, leading to a large variety of different turbulent combustions models. Many state-of-the-art turbulent combustion models employed in LES make stringent key assumptions, for instance on the regime-specific flame-structures (e.g., laminar flamelet models [13]), molecular diffusion (e.g., transported probability density function models [14]), and highly simplified turbulent mixing (e.g., linear eddy model [15] and one-dimensional turbulence model [16]). The current study is part of a longer term effort to improve LES accuracy for prediction of flame-turbulence interactions in premixed turbulent combustion systems by incorporating directly key SFS features that significantly impact the evolution of RS motions through key nonlinear dynamic couplings.

A. Research Strategy and Organization

In the current study we explore the potential of new modeling elements embedded within current state-of-the-art LES framework to capture dynamically dominant inter-scale couplings between RS and SFS necessary for more accurate prediction of the evolution of RS premixed turbulent combustion. Figure 1 illustrates the strategy upon which the current study is based. To mathematically represent the dynamical interactions among the turbulence length scales of the key dynamical variables, it is necessary to represent the nonlinear terms in the dynamical system within a scale-based decomposition consistent with the LES framework. The Fourier basis has been historically used for this purpose [17–19] and has important mathematical and representational advantages of which we take advantage in the current study. Furthermore, the multi-dimensional Fourier spectral decomposition may be regarded as the most "elemental" in the sense that other decompositions may be represented as filtered versions of the Fourier decomposition (e.g., wavelet decomposition [20]). However, the Fourier representation is often regarded as only useful for periodic signals, and therefore for turbulence systems that are statistically homogeneous in all spatial directions. We show in the current study that this perception is incorrect; we show here that correlations can be made between spatial "coherent structure" and spectral "coherent structure" even in directions that are statistically inhomogeneous.

However, to do so there is a practical issue that must be overcome, illustrated by "Stage 1" in Fig. 1 where the physical-space signals are transformed to Fourier space. Because the Fourier transform of analytic functions on finite domains mathematically represent periodic extensions of the signal in all coordinate directions, in non-periodic directions the Fourier representation includes the discontinuity in the signal at the boundaries between the signal and its periodic extension. To use Fourier space quantitatively in inhomogeneous directions, as we do, it is necessary to remove the spurious Fourier spectral content associated with the boundary discontinuity in those directions. This is referred to as "signal discontinuity pollution removal" in Stage 1 of Fig. 1. Once done, the kinematic relationships between physical space coherent structure in key dynamic variables in premixed flame-turbulence interaction and their Fourier spectral content must be analyzed and characterized in context with the LES decomposition into grid-resolved and subfilter scales (Stage 2 in Fig. 1). Finally, the inter-scale interactions between key kinematic structures must be identified and quantified. Here we take advantage of the mathematical simplicity of the Fourier spectral description of the equations of motion where quadratic nonlinearities attain a "triadic form" whereby each elemental triadic interaction occurs among three Fourier modes whose wave-vectors form a triangle in multidimensional Fourier space. We take advantage of this elemental representation of key nonlinearities in order to arrange and down select the dominant triadic couplings between SFS and RS motions, and the corresponding SFS content associated with dominant RS-SFS dynamics (Stage 3 in Fig. 1).

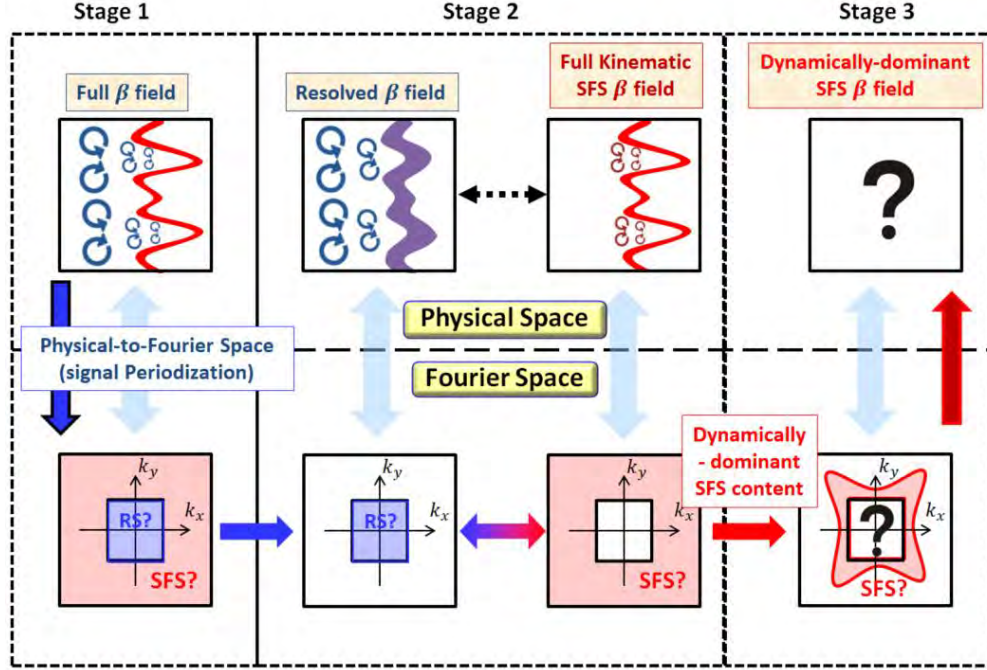


Fig. 1 Schematic of overall research strategy.

Each of the stages illustrated in Fig. 1 are the subject of three separate manuscripts [21, 22]. The current study addresses “Stage 3”, the development of a procedure to identify dynamically dominant inter-scale couplings and analysis of the dominant SFS content within these couplings for application to LES of premixed turbulent combustion. The specific objectives are threefold. Firstly, we use the previous framework develop in Stages 1 and 2 [21, 22] to relate spectral content to physical space content of key variables in premixed turbulent combustion. Secondly, we use this knowledge to characterize nonlinear triadic inter-scale couplings between RS and SFS. Finally, we identify dynamically dominant inter-scale couplings and extract the SFS content within these couplings.

This manuscript is organized as follows. The general characteristics and mathematical framework of inter-scale couplings in physical and Fourier space are presented in Sec. II. The design of the reduced-physics simulations consisting of flame-vortex interactions is discussed in Sec. III. The Fourier-physical space relationships of key variables in context with the LES framework identified (Stage 2 [22]) is summarized in Sec. IV. Sec. V presents the method to identify the dynamically dominant inter-scale couplings, extract the associated SFS content within the couplings, and analyze its Fourier and physical space characteristics. The key findings of this study are summarized in Sec. VI.

II. Characteristics of Key Nonlinearities in Physical and Fourier Space

As an inherently nonlinear high degree-of-freedom dynamical system, the interactions among scales of motion in premixed turbulent combustion are central both to the dynamics and simulation/modeling of flame-turbulence interactions. In this paper we focus on the dynamical interactions among turbulence scales through the advective nonlinearities in the filtered governing equations of momentum, enthalpy, and species transport within LES frameworks. The structure of the advective nonlinearity for the evolution of the filtered scales of motion resolved on an LES grid can be written as follows:

$$\left. \frac{\partial \vec{\beta}^r(\vec{x})}{\partial t} \right|_{adv} = -\nabla \cdot (\vec{\beta}^r \vec{u}^r)^r - \nabla \cdot (\vec{\beta}^r \vec{u}^s + \vec{\beta}^s \vec{u}^r + \vec{\beta}^s \vec{u}^s)^r, \quad (1)$$

where $\vec{u}$ is the velocity vector, $\vec{\beta}$ represents primitive dependent variables on a per unit volume basis (momentum $\rho \vec{u}$, mixture enthalpy ρh , and species mass concentration ρY_i), superscripts r and s imply variables filtered at resolved and subfilter scale, respectively, and the temporal dependence of $\vec{u}$ and $\vec{\beta}$ is omitted for simplicity. The first term in the

RHS of Eq. (1) represents the inter-scale nonlinear advective couplings fully-resolved in the simulation on the LES grid and referred as “resolved-scale-resolved-scale” (RS-RS) couplings. This should be the leading-order term in the equation of motion for $\tilde{\beta}^r$ if the grid is properly designed under the fundamental LES assumptions discussed in Sec. I. The second term in the RHS of Eq. (1) describes the inter-scale couplings between SFS and RS motions, which we refer as “resolved-scale-subfilter-scale” (RS-SFS) couplings. Under the LES framework, this term must be modeled but should be higher order in Eq. (1).

We must analyze the dynamical interactions among scales represented by Eq. (1) in a scale-space view where nonlinear couplings between scales are explicitly represented. Fourier-spectral space is particularly useful for this purpose, in that “scales of motion” are defined with a complex (exponential) basis of elemental spectral content each of which represents precisely-defined scale, direction, and phase. Most importantly, advective nonlinearities describing interactions among scales of motion appear explicitly in the transformed governing equations in a geometrically straightforward “triadic” manner [17, 18].

The structure of the advective nonlinearity for the evolution of the resolved scales in Fourier space is given by the Fourier transform of Eq. (1) as

$$\left. \frac{\partial \tilde{\beta}(\vec{k}^r)}{\partial t} \right|_{adv} = \sum_{\vec{p}_\Delta} \tilde{\beta}(\vec{p}_\Delta) i \vec{k}^r \cdot \tilde{u}(\vec{q}) = \sum_{\vec{p}_\Delta} \hat{\Delta}(\vec{k}^r, \vec{p}_\Delta; \vec{q}), \quad (2)$$

where $i = \sqrt{-1}$, hats ($\hat{\cdot}$) indicate Fourier coefficients, $\vec{k}^r$ is a wave-vector at the resolved scales, and subscript Δ implies that a “triad” wave-vector $\vec{q}$ is involved in each term such that $\vec{k}^r$, $\vec{p}_\Delta$, and $\vec{q}$ form a triangle $\vec{q} = \vec{k}^r - \vec{p}_\Delta$, illustrated in Fig. 2. Equation 2 shows that the dynamical evolution of a single RS Fourier mode $\tilde{\beta}(\vec{k}^r)$ due to nonlinear advection is given by the sum of all “triadic” interactions involving Fourier modes $\tilde{\beta}(\vec{p}_\Delta)$ and $\tilde{u}(\vec{q})$, defined by $\hat{\Delta}(\vec{k}^r, \vec{p}_\Delta; \vec{q})$. As such, the time derivative of $\tilde{\beta}(\vec{k}^r)$ lives in the resolved scales subregion and evolves through interactions with other turbulence scales of motion through a large sum of inter-scale interactions.

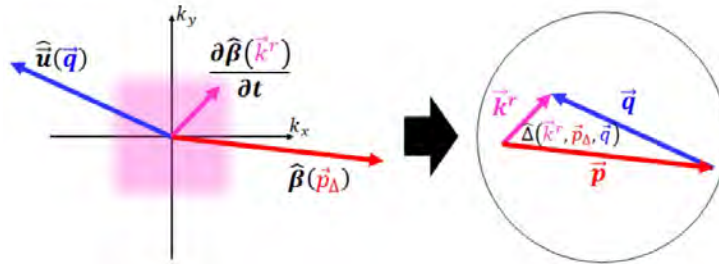


Fig. 2 Fundamental triadic form of the advective nonlinearity in Eq. (2). The advective nonlinear contribution to the time evolution of $\tilde{\beta}(\vec{k}^r)$ ($\tilde{\beta} = \rho \tilde{u}$, ρh , and ρY_{HCO}) is given by the summation of all triads where each triad represents the fundamental interaction between wave-vectors $\vec{k}^r$, $\vec{p}_\Delta$, and $\vec{q} = \vec{k}^r - \vec{p}_\Delta$ in multidimensional Fourier space, representing $\hat{\Delta}(\vec{k}^r, \vec{p}_\Delta; \vec{q})$.

Within the summation in the RHS of Eq. (2) the fully-resolved triadic interactions are identified as $\hat{\Delta}(\vec{k}^r, \vec{p}_\Delta^r; \vec{q}^r)$, while the RS-SFS interactions are given by all triads with at least one SFS wave-vector, $\hat{\Delta}(\vec{k}^r, \vec{p}_\Delta^r; \vec{q}^s)$, $\hat{\Delta}(\vec{k}^r, \vec{p}_\Delta^s; \vec{q}^r)$, or $\hat{\Delta}(\vec{k}^r, \vec{p}_\Delta^s; \vec{q}^s)$. The dynamical evolution of the resolved scales in physical space due to the advective nonlinearity (Eq. (1)) is recovered by projecting Eq. (2) back to physical space using the Fourier basis,

$$\left. \frac{\partial \tilde{\beta}^r(\vec{x})}{\partial t} \right|_{adv} = \sum_{\vec{k}^r} \left. \frac{\partial \tilde{\beta}(\vec{k}^r)}{\partial t} \right|_{adv} e^{i \vec{k}^r \cdot \vec{x}} = \sum_{\vec{k}^r} \sum_{\vec{p}_\Delta} \hat{\Delta}(\vec{k}^r, \vec{p}_\Delta; \vec{q}) e^{i \vec{k}^r \cdot \vec{x}}. \quad (3)$$

The condition that the LHS of Eq. (3) is real (“reality condition”) creates symmetry in Fourier space with respect to the wavenumber origin, so that $\hat{\phi}(-\vec{k}^r) = \hat{\phi}^*(\vec{k}^r)$ for an arbitrary real-valued function $\hat{\phi}$, where superscript $*$ implies

complex conjugate. Thus, the wave-vector space is separable into two subsets $\vec{k}^r = [-\vec{k}_+^r, \vec{k}_+^r]$, where $\vec{k}_+^r$ implies positive set $\vec{k}^r > 0$, and the projection of Eq. (3) may be rewritten over $\vec{k}_+^r$ as,

$$\left. \frac{\partial \vec{\beta}^r(\vec{x})}{\partial t} \right|_{adv} = \sum_{\vec{k}_+^r} \frac{\partial \widehat{\vec{\beta}}(\vec{k}_+^r)}{\partial t} e^{i\vec{k}_+^r \cdot \vec{x}} + \sum_{-\vec{k}_+^r} \frac{\partial \widehat{\vec{\beta}}(-\vec{k}_+^r)}{\partial t} e^{-i\vec{k}_+^r \cdot \vec{x}} = \sum_{\vec{k}_+^r} \frac{\partial \vec{\beta}_{sw}(\vec{x}; \vec{k}_+^r)}{\partial t}, \quad (4)$$

where

$$\frac{\partial \vec{\beta}_{sw}(\vec{x}; \vec{k}_+^r)}{\partial t} = 2 \left\{ \text{Re} \left[\frac{\partial \widehat{\vec{\beta}}(\vec{k}_+^r)}{\partial t} \right] \cos(\vec{k}_+^r \cdot \vec{x}) - \text{Im} \left[\frac{\partial \widehat{\vec{\beta}}(\vec{k}_+^r)}{\partial t} \right] \sin(\vec{k}_+^r \cdot \vec{x}) \right\}. \quad (5)$$

We refer to the form written in Eq. 5 as the “shear wave” form in which a point in Fourier space is written in physical space, indicated by subscript $_{sw}$ [23] (the subscript $_{adv}$ is not shown on the RHS for simplicity). The operators Re and Im in Eq. (5) imply the real and imaginary parts of a complex number and $\vec{k}^r = 0$ is not included in this derivation as it represents the volume-averaged value, which does not have scale content. In Eq. 5, the physical space structure of a Fourier coefficient $\partial \widehat{\vec{\beta}}(\vec{k}_+^r)/\partial t$ with its symmetric pair at $-\vec{k}_+^r$ represents a physical-space “shear wave”, $\partial \vec{\beta}_{sw}(\vec{x}; \vec{k}_+^r)/\partial t$, with specific direction, length scale, amplitude, and phase. This concept is used to obtain the physical-space counterpart of Eq. (2) for a Fourier mode at a point in the positive subset of the RS wave-vector space,

$$\frac{\partial \vec{\beta}_{sw}(\vec{x}; \vec{k}_+^r)}{\partial t} = \sum_{\vec{p}_\Delta} \vec{\Delta}_{sw}(\vec{x}; \vec{k}_+^r, \vec{p}_\Delta, \vec{q}), \quad (6)$$

so that Eq. (3) becomes

$$\left. \frac{\partial \vec{\beta}^r(\vec{x}, t)}{\partial t} \right|_{adv} = \sum_{\vec{k}_+^r} \left. \frac{\partial \vec{\beta}_{sw}(\vec{x}; \vec{k}_+^r)}{\partial t} \right|_{adv} = \sum_{\vec{k}_+^r} \sum_{\vec{p}_\Delta} \vec{\Delta}_{sw}(\vec{x}; \vec{k}_+^r, \vec{p}_\Delta, \vec{q}), \quad (7)$$

where the reality condition is also employed for triad pairs $\widehat{\vec{\Delta}}(-\vec{k}_+^r, \vec{p}_\Delta; \vec{q}) = \widehat{\vec{\Delta}}^*(\vec{k}_+^r, -\vec{p}_\Delta, -\vec{q})$ to obtain the shear wave triads in physical space $\vec{\Delta}_{sw}(\vec{x}; \vec{k}_+^r, \vec{p}_\Delta, \vec{q})$ (Appendix B).

The symmetry in Fourier space created by the reality condition indicates that only half of the wave-vectors $\vec{k}^r$ need to be analyzed for the evolution of the resolved scales. However, all wave-vectors $\vec{p}_\Delta$ need to be included in the Fourier space representation of the advective nonlinearity for a given $\vec{k}^r$ as they represent distinct individual inter-scale couplings (RHS of Eq. (7)). To simplify the writing, we omit the subscripts $_+$ in the remainder of this manuscript since the choice of half of the wave-vectors $\vec{k}^r$ is arbitrary.

The mathematical framework developed here provides a description to identify and quantify inter-scale interactions between key kinematic structures through the advective nonlinearities in the filtered governing equations. In future research we will apply this methodology to analyze other key nonlinearities associated with finite-rate chemistry, heat release, and pressure-dilatation work in the governing equations. Using this framework, the objectives of this work are: (1) identify the dynamically dominant triadic interactions in Eq. (6) in the evolution of resolved-scale momentum, enthalpy, and species concentration (all per unit volume) and (2) isolate the SFS content within the dynamically dominant triadic couplings in Fourier and physical space. The procedure to downselect the dynamically dominant triadic interactions is presented in V.

III. Simulation Configuration and Numerical Details

In this section, key non-dimensional parameters used in the design of the numerical simulation are described along with details of computational methodology.

A. Reduced Physics Simulations: Flame-Vortex Interactions

In turbulent premixed flames, stiff nonlinear chemical reactions involving many reacting scalars introduce broad ranges of length and time scales that overlap with the characteristic scales of turbulence motions, resulting in strong two-way couplings between turbulence and combustion [24–26]. These scales are typically organized in the form of key

non-dimensional numbers within a parameter space that characterizes the regimes of turbulent premixed combustion, as proposed by Borghi [24], Peters [25], and Williams [26]. A key non-dimensional parameter that characterizes these regimes is the Karlovitz number,

$$Ka \equiv \tau_F / \tau_\eta \sim \gamma (u_\eta / s_L)^2 \sim \gamma^{-1} (l_F / \eta)^2, \quad (8)$$

where $\tau_F = l_F / s_L$ is the flame time scale (s_L is the laminar flame speed and l_F is the laminar flame thickness), $\tau_\eta = \eta^2 / \nu$ is the Kolmogorov time scale (u_η and η are the Kolmogorov velocity and length scales) and ν is the kinematic viscosity. In Eq. (8), $\gamma \equiv s_L l_F / \nu \approx s_L l_F / \alpha \sim O(1)$, when the Prandtl number $\nu / \alpha \approx 1$ (as is typical of gases), when ν is the kinematic viscosity and α is the thermal diffusivity of the fluid in the reaction zone. Classical scaling arguments [24–26] suggest that when $Ka \ll 1$, $1 \lesssim Ka \lesssim 10^2$, and $Ka \gg 10^2$ the “flamelet”, “thin reaction zone”, and “broken or distributed reaction zone” regimes, respectively, are achieved.

Some key fundamental dynamics of flame-vortex interactions may be conceptualized within reduced-physics systems that characterize of local interactions between a premixed flame and single-scale turbulence eddies from within a high Reynolds number turbulent reacting flow [27], as supported by recent experimental visualizations [28]. Using this framework, single-scale eddies in the inertial sub-range of a turbulent flow in the unburnt mixture are represented by vortices characterized by its length scale, l_v'' , and velocity scale, u_v'' . These vortex scales are typically normalized and organized in the parameter space referred to as the spectral diagram [27], where the Karlovitz number can be expressed in terms of vortex scales by applying classical Kolmogorov scaling to Eq. (8), as

$$Ka \sim (l_v / \delta_f)^{-1} (u_v / s_L) (Re_v)^{1/2}, \quad (9)$$

where $Re_v = u_v l_v / \nu$ is the vortex Reynolds number in the unburnt mixture.

In this work, we employ two-dimensional (2D) flame-vortex interactions as a cost-effective configuration with highly reduced dynamic complexity to provide important insight regarding relationships between physical space coherent structure in key dynamic variables in premixed flame-turbulence interaction and their Fourier spectral content. This simplified configuration captures key hydrodynamics and chemistry kinetics effects and have been successfully used in previous studies to investigate premixed turbulent combustion regimes [27, 29], vorticity dynamics [30, 31], pocket formation [32], flame stretch [33], and flamelet quenching conditions [27, 34, 35] in turbulent premixed flames.

B. Problem Setup

The numerical simulation was performed using a 2D rectangular domain illustrated in Fig. 3 with length $L_y = 4D$ and $L_x = 16D$ in the cross-stream and streamwise directions, respectively, where D is the vortex diameter. An inflow of unburnt *n*-heptane/air mixture with $\phi = 0.95$, $T_u = 300$ K, and $P = 1$ atm is injected in the streamwise direction, the burnt products exit through the outflow plane, and periodic boundary condition is enforced in the cross-stream direction. The thermochemical conditions in the unburnt mixture are selected according to those used in the experiments of moderate Ka turbulent premixed flames [36].

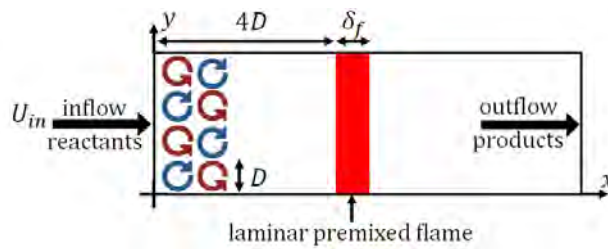


Fig. 3 Schematic of computational domain for simulation of 2D flame-vortex interactions.

A planar *n*-heptane/air laminar premixed flame with the same unburnt conditions is initially placed 4 vortex diameters away from the inflow boundary. The planar laminar flame thickness and flame burning speed are computed to be 0.5 mm and 39 cm/s, respectively, using the chemical kinetic models described in Sec. III.C. The laminar flame thickness is defined as the distance between the iso-levels of 5% and 85% of progress variable, $C = (T - T_u) / (T_b - T_u)$, where T_u and T_b are the temperatures in the unburnt and burnt mixture, respectively.

An array of counter-rotating vortex pairs is continuously injected in the inflow boundary using time varying Dirichlet boundary conditions for the velocity components. The array of vortices is kinematically constructed from a combination

of two sine waves aligned with the cross-stream and streamwise directions resulting in an array of single-scale vortices with zero averaged velocity. A streamwise injection velocity U_{in} is added to the kinematic velocity field, such that U_{in} is set to match the statistically averaged flame burning speed from the simulation. The same vortices that are injected in the inflow boundary are placed in the reactants side of the computational domain ($x \leq 4D$) as initial condition for the simulation.

The vortex length scale, l_v , is computed as the square root of the surface area within the iso-level of 30% of the maximum vorticity divided by $\pi/4$, which captures more than 90% of the total vortex circulation. As such, l_v represents the effective diameter of the vortex core where most of the circulation is contained (63% of the vortex diameter in the sinusoidal velocity distribution described above, $l_v = 0.63D$). Accordingly, the vortex velocity scale, u_v , characterizes the strength of the vortex core, and is computed as the circulation contained within the same vorticity iso-level divided by its arclength.

The simulation is carried out employing vortices representative of inertial-range eddies in the reactant side of turbulent premixed flames in the thin reaction zone regime ($Ka \sim o(1)$). The vortex length scale is significantly larger than the flame length scale ($l_v/\delta_f = 10$) to elucidate the distinct characteristics of vortices and the premixed flame front in scale (Fourier) space. Therefore, the simulation is design such that $l_v/\delta_f = 10$, $u_v/S_L = 0.5$, $Ka = 2.5$, and $Re_v = 250$, where the reported values are computed after the vortices travel from the inflow plane to the vicinity of the flame front on the unburnt side.

C. Computational Methodology

The 2D flame-vortex interaction simulations were performed using the NGA code [37], where unsteady Navier-Stokes equations under the low Mach number approximation and the transport equations for temperature and mass fractions of the various species are solved simultaneously. The scalar equations were discretized using the BQUICK scheme, which ensures that the physical bounds of appropriate quantities are numerically preserved throughout the simulation without adding significant artificial diffusion [38]. A recently-developed computationally-efficient, semi-implicit, iterative method is used for the time-integration of chemical source terms of gas-phase species [39]. This method has been shown to be free of lagging errors, as efficient as an explicit time-integration per time step, and capable of using large, stable time step sizes [40].

A semi-detailed chemical kinetic model containing 35 species and 217 reactions (forward and backward reaction counted separately) [41] is used. This mechanism was reduced from the originally-developed full mechanism with 149 species and 1651 reactions [42]. The reader is referred to [41, 43] for complete details about the chemistry reduction procedure and chemical model validation. This chemical model has been used in moderate Ka turbulent premixed flame simulations to reduce the computational cost [41].

The computational mesh is non-uniform in streamwise direction and uniform in cross-stream direction, being most refined in the vortex and flame regions in streamwise direction and stretched towards the outflow boundary. At least 80 grid points are used to resolve the vortex length scale in the reactants side and 20 grid points within the flame front thickness. The numerical simulation is shown to give the same results when twice as many grid points are used in both directions and the time step size is $1 \mu s$ to accurately resolve the chemical time scales.

D. Removing “Pollution” from Boundary Discontinuity of Non-Periodic Signals

The details of the summary presented here are given in [21]. The Fourier transform of physical-space signals over finite domains mathematically represents periodic extensions of the physical-space signal in all coordinate directions, as the Fourier basis functions are periodic and defined over an infinite domain. In non-periodic directions, the Fourier representation includes the physical-space discontinuity in the signal at the boundaries between the signal and its periodic extension. The boundary discontinuities in physical space result in a broadband power-law distribution of Fourier energy over all wavenumbers, which may overlap with the scale content of frontal structures of interest within the domain (e.g. flame fronts) characterizing pollution in Fourier space. This pollution must be removed in order to investigate relationships between physically-relevant coherent structure in Fourier and physical space (Stage 2 in Fig. 1), as we do here, and characterize inter-scale interactions between key kinematic structures (Stage 3 in Fig. 1).

To use Fourier space quantitatively in non-periodic directions, we applied a recently developed “signal discontinuity pollution removal procedure” [21] to all variables of interest with $\alpha = 0.85$ and considering only C^0 boundary discontinuities. As a result, the maximum signal modification parameter and residual boundary discontinuity are smaller than 5% and 30%, respectively. The reader is referred to the original manuscript [21] for more details regarding the procedure and only a brief overview is provided here and in Appendix A.

To illustrate the outcome of the procedure, Fig. 4 presents the original and post-processed signals in Fourier space of ρu analyzed in Sec. IV.C. We plot iso-contours of the logarithmic squared Fourier coefficients and the color bars represent logarithmic values spanning 5 orders of magnitude. The wavenumber axes are normalized using the planar laminar flame thickness. Our analysis focuses in Fourier modes associated with scale down to the flame scale, that is scales within the flame-scale box defined by $k_{x,y}\delta_f/\pi = \pm 1$. The pink box identifies the vortex scale defined by $k_{x,y}\delta_f/\pi = \pm\delta_f/D = 0.06$, where π/D is the vortex-scale wavenumber associated with the four forcing Fourier modes in the kinematically constructed array of vortices. The forced even spanwise symmetry of vortices leads to negligible Fourier content in Fourier modes odd in k_y (i.e., at $k_y/\Delta k_y = \pm 1, \pm 3, \dots$), which are removed from the iso-contour plots for better visualization.

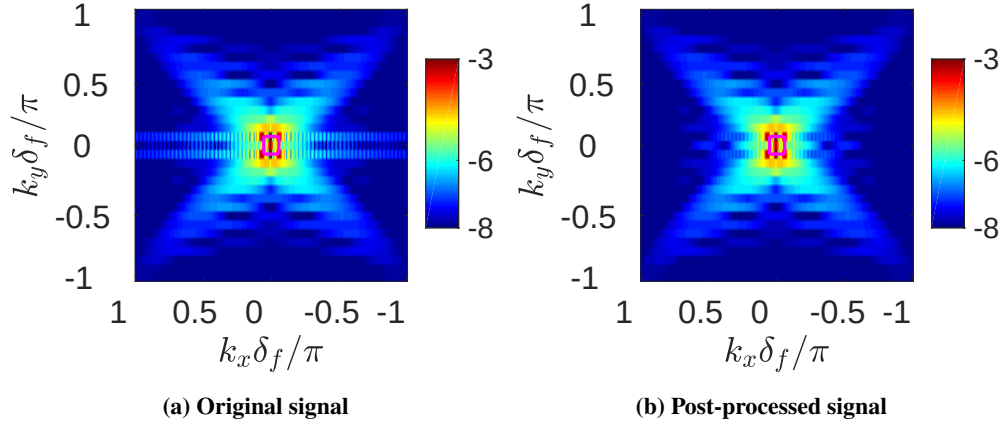


Fig. 4 Iso-contours of the logarithmic squared Fourier coefficients $\log_{10}(\widehat{\rho u}^2)$ of (a) original and (b) post-processed signal.

The unphysical power-law pollution associated with boundary discontinuities are observed in Fig. 4a as “bands” over k_x with large Fourier content at $|k_y|\delta_f/\pi \leq 0.06$ (i.e., at $k_y/\Delta k_y = 0, \pm 2$), which are associated with the inflow of vortices in the reactants and outflow of elongated streaks in the products side of the mixture (Sec. IV.B). Figure 4b shows that the procedure effectively removes the spurious pollution over these bands with minimal modification at Fourier modes within the vortex-scale box.

IV. Summary of Structural Relationships between Physical and Fourier space in Flame-Vortex Interactions

In this chapter we summarize results relevant to the current study that was described in detail in [22]. To better understand the relationships between physical-space and Fourier-space coherent structural features from the flame-vortex simulation described in Sec. III, we use knowledge from “Stage 2” [22] (Fig. 1) of Fourier-physical relationships of key variables to characterize nonlinear triadic inter-scale couplings between RS and SFS in the Sec. V. Four primary variables are chosen to represent the key features of the 2D flame-vortex interactions: the components of the momentum vector (ρu and ρv) as representative of hydrodynamic effects; the mixture enthalpy (ρh) as representative of thermal energy; and the formyl radical mass per unit volume (ρY_{HCO}) representing an intermediate species that identifies heat release rate by combustion [44]. We also analyze the components of the velocity vector (u and v), which form the advective nonlinearity in the governing equations of each primary variable (Sec. II). A single time instant within the statistically stationary state of the simulation is selected for analysis, when the flame is most corrugated based on the largest difference between the maximum and minimum streamwise location of the progress variable iso-level, $C = 0.5$. The statistically stationary state is observed after a transient period from the initial condition at approximately 4 vortex advection time scales (D/U_{in}).

A. Defining “Resolved” LES Scales

For a consistent interpretation of our reduced-physics simulations in the view of LES, we developed a procedure [22] to decompose scales into resolved and subfilter scale based on the fundamental LES criterion described in Sec. I. As such, the “resolved scale” motions should contain “most” of the integral-scale motions in momentum and energy (or

temperature), where “most” is generally 80-90%. In Stage 2 [22] we defined the spectral RS cutoff between resolved and subfilter scale motions such that at least 85% of the variance of ρu , ρv , and ρh are captured at the resolved scales. Variance is calculated after removing the two-dimensional averaged value, which is equivalent to the sum of squared Fourier coefficients excluding $\vec{k} = 0$ in Fourier space (Parseval).

The “resolved scales” are defined using a 2D sharp low-pass rectangular filter in Fourier space with wavenumber cutoffs $k_{RS,x}\delta_f/\pi = 0.1$ and $k_{RS,y}\delta_f/\pi = 0.26$, with corresponding smallest resolved length scale in streamwise and cross-stream directions $l_{RS,x} = \pi/k_{RS,x} = 0.62D$ and $l_{RS,y} = \pi/k_{RS,y} = 0.24D$, respectively. The procedure does not include the variance content of ρY_{HCO} , which was observed [22] to characterize heat release at the flame scales that cannot generally be resolved in practical LES (only 8% of the variance for ρY_{HCO} is captured at the resolved scales). We observed that key combustion intermediates such as ρY_{HCO} are greatly not resolvable by the LES grid, which deviates from the basic LES requirement so that not all dynamical equations for turbulent reacting flows can be treated using classical LES framework and associated SFS modeling strategies. For this reason, in this work we investigate and contrast the dynamically dominant RS-SFS inter-scale interactions of key combustion intermediates (ρY_{HCO}), thermal energy (ρh), and momentum components (ρu and ρv) (Stage 3 in Fig. 1).

B. Analysis of Resolved and Subfilter Scale Structure: Physical Space

The RS filter described above is used to decompose RS and SFS content in both physical and Fourier space for the four primary variables and the associated velocity components analyzed in this work. Figure 5a-f shows the 2D instantaneous iso-contours of streamwise and cross-stream components of velocity (u and v) and momentum (ρu and ρv) vectors, the mixture enthalpy (ρh), and formyl concentration (ρY_{HCO}), respectively. In these figures, the bottom and top halves show the original and RS filtered signals, where the two solid curves indicate the RS flame front defined by RS progress variable iso-levels of 5% and 85%.

Figure 5 shows that 2D vortices enter the reactants side of the mixture from the inflow boundary conditions and interact with the flame causing flame distortions symmetrically in y . Over each interaction, flame cusps are formed over regions where the induced velocity of two adjacent vortices in the streamwise direction is towards the burnt products (symmetrically at $y/D = 1$ and 3 in Fig. 5). We make use of this symmetry to show the resolved and original signals in the top and bottom half, respectively, of each iso-contour in Fig. 5. The vortices elongate in the streamwise direction as they cross the flame due to flow acceleration and volumetric expansion within the flame front, resulting in increase of streamwise velocity across the flame front (Fig. 5a). When compared to the velocity components, the momentum components include the effect of density reduction from reactants to product side of the mixture as result of increase in temperature (Fig. 5a-d). The change in the sign of v and ρv across the flame is a consequence of Flame Generated Vorticity (FGV) by baroclinic torque [31, 35] that changes the sign of vortices as they cross the flame front. In contrast with momentum components, ρh and ρY_{HCO} in Fig. 5e-f are approximately spatially uniform on the reactants and products sides of the flame with most of its variations within the thin flame region. ρh has a jump from reactants to products, while ρY_{HCO} is a combustion intermediate that is highly localized and peaks within the flame, in the reaction zone.

From the qualitative description above, four distinct physical-space coherent structural features can be identified in the key variables: vortices on the reactants side and elongated structures (“streaks”) on the products side in momentum and velocity, a corrugated flame front in all variables, and the ensemble mean streamwise front in streamwise velocity components, enthalpy, and formyl concentration that characterizes the overall change from reactants to products. The large scale structural features of key variables are readily identified in the RS signals in the top halves of Fig. 5. These include the vortices in the reactants side and the elongated streaks on the products side for momentum and velocity components (Fig. 5a-d). The RS corrugated flame front is less corrugated and thicker, with corresponding lower flame-normal gradients within the front, than the full field which contains the small-scale features with the SFS motions.

C. Analysis of Resolved and Subfilter Scale Structure: Fourier Space

To better understand how the information regarding the physical-space coherent structures described in Sec. IV.B is organized in collections of Fourier coefficients characterizing Fourier-space coherent structure, systematic filters are employed in Stage 2 [22] and we summarize the results here.

Figure 6 presents the concurrent view in Fourier space of the physical-space signals shown in the bottom halves of Fig. 5. We plot iso-contours of the logarithmic squared Fourier coefficients for each variable and the color bars represent logarithmic values spanning 5 orders of magnitude. The squared Fourier coefficients are symmetric with respect to the wavenumber origin due to the “reality condition” [22] (i.e., $\hat{\phi}^2(-\vec{k}) = \hat{\phi}^2(\vec{k})$ for Fourier coefficients $\hat{\phi}$).

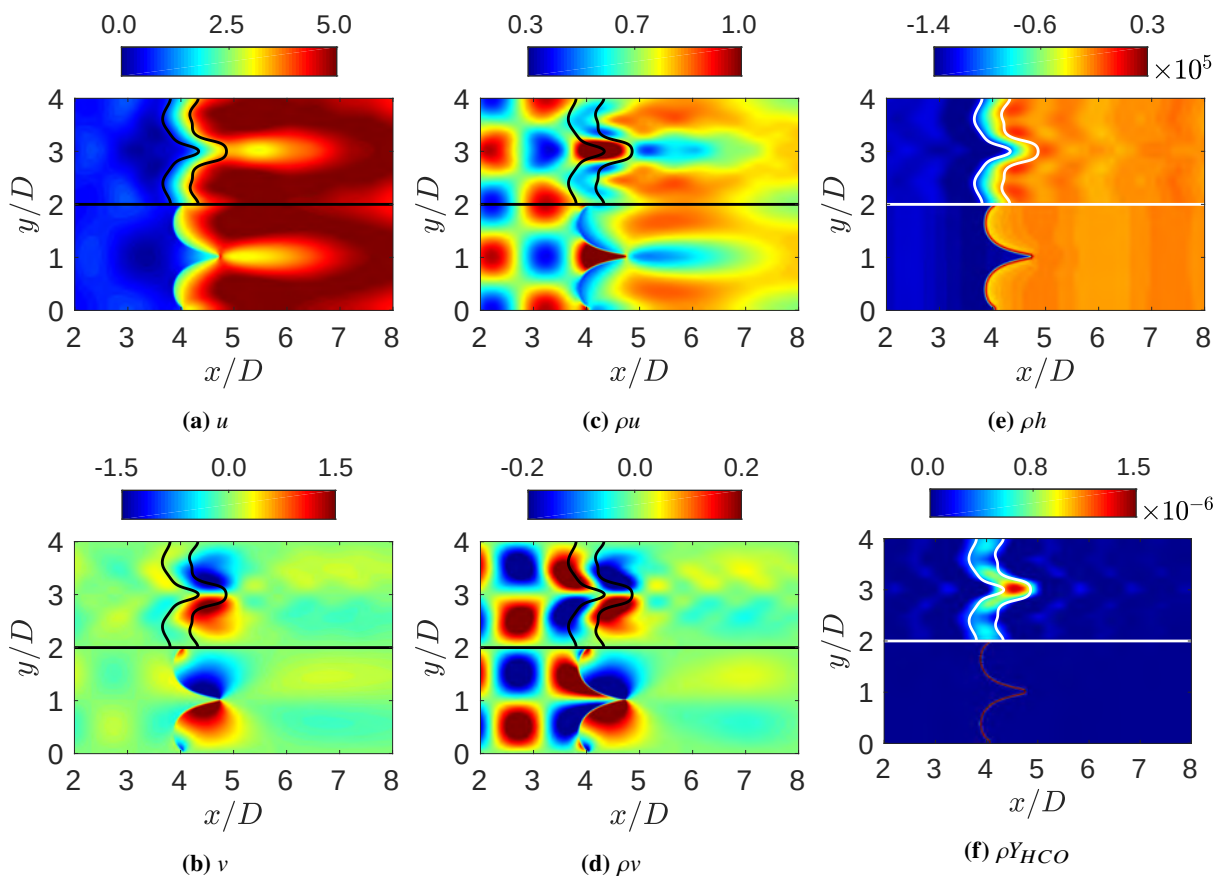


Fig. 5 Iso-contours of (a-d) streamwise and cross-stream components of velocity (u and v in m/s) and momentum (ρu and ρv in kg/m^2s) vectors, (e) mixture enthalphy (ρh in J/m^3), and (f) formyl concentration (ρY_{HCO} in kg/m^3), respectively. Resolved and original signals are shown in the top and bottom half of each figure, respectively.

An additional symmetry arises over k_y from the forcing with the array of vortices that are symmetric in the cross-stream direction, so that $\hat{\phi}^2(k_x, k_y) \approx \hat{\phi}^2(k_x, -k_y)$. In addition, the forced even spanwise symmetry leads to negligible Fourier content in Fourier modes odd in k_y (i.e., at $k_y/\Delta k_y = \pm 1, \pm 3, \dots$), which are removed from all iso-contour plots for better visualization of structural features. In Fig. 6a-b we make use of these symmetries to show the squared Fourier coefficients of the streamwise and cross-stream components of momentum and velocity vectors in the top and bottom halves of each figure, respectively.

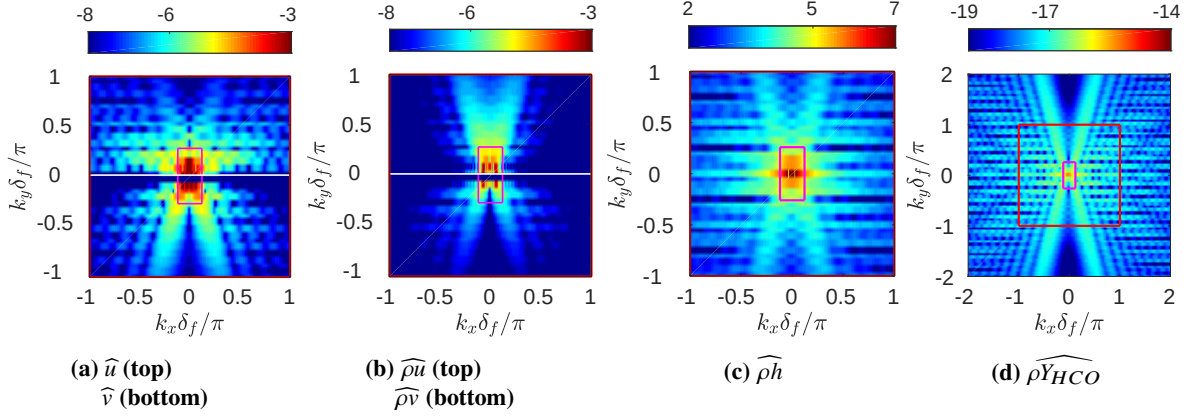


Fig. 6 Iso-contours of the logarithmic squared Fourier coefficients of key variables in Fourier space for the flame-vortex interaction shown in Fig. 5. (a) $\log_{10}(\widehat{u}^2)$ (top) and $\log_{10}(\widehat{v}^2)$ (bottom); (b) $\log_{10}(\widehat{\rho u}^2)$ (top) and $\log_{10}(\widehat{\rho v}^2)$ (bottom); (c) $\log_{10}(\widehat{\rho h}^2)$; (d) $\log_{10}(\widehat{\rho Y_{HCO}}^2)$.

The wavenumber axes in Fig. 6 are normalized using the planar laminar flame thickness so that the flame-scale box is defined by $k_{x,y}\delta_f/\pi = \pm 1$ (solid red lines in Fig. 6). The solid magenta box towards small wavenumbers identifies the RS cutoff scales defined in Sec. IV.A. For the key variables shown in Fig. 6, the highest magnitude squared Fourier coefficients are found to be organized within a “star-like” pattern towards large wavenumbers (i.e. small scales) that extends from the vortex scale box to the flame scale box for velocity, momentum, and enthalpy. For ρY_{HCO} , energetic Fourier content was observed to extend up to $k_{x,y}\delta_f/\pi \approx \pm 3$ (not shown here), which is associated with the reaction zone thickness of the premixed flame. As discussed in our previous works [21, 22], the star-like patterns in Fourier space are, in reality, power-law variation arising from near-discontinuities at the flame front. Given the finite thickness of the flame front and the sharp, but smooth, variation across the front, the squared Fourier coefficients exhibit power-law decay up to the flame front thickness. At the flame thickness, the squared Fourier coefficients transition to exponential decay to the grid cutoff scale. The directions of the “star legs” are normal to the corresponding sections of flame front that generates the power-law decay.

From the perspective of typical LES scale decomposition, the corrugated flame front is a physical-Fourier space coherent structure that resides partially at resolved and subfilter scales. As such, one would expect dominant inter-scale couplings between RS and SFS among Fourier modes associated with this coherent structure, as it will be shown next in Sec. V. The modeling of inter-scale couplings are more critical for combustion intermediates directly involved in heat release, such as ρY_{HCO} , in the current dynamical system where variance content is almost entirely at the subfilter scales.

V. Dynamically Dominant Inter-Scale Couplings and Subfilter-Scale Content

LES imposes a dramatic reduction in degrees of freedom in the discretized dynamical system by restricting explicit predictions to a very small number of “resolved scale” modes relative to the total number of modes required to evolve the dynamical system up to the smallest dynamically relevant scales of motion. In this reduced system, the dynamical effects of the unresolved SFS motions on the evolution of the RS motions require modeling. The evolution of the RS motion is dynamically coupled to all other scales of motion through nonlinear terms in the filtered governing equations, such as the advective nonlinearity in Eq. (1). These terms must be modeled within the LES framework wherein only RS data on the LES grid are available, in principle. As described in Sec. II, our goal is to identify *dynamically* dominant inter-scale couplings. By grouping these triadic terms according to specific features, we extract the corresponding SFS content of $\tilde{\beta}$ and $\tilde{u}$ associated with dynamically dominant RS-SFS couplings. Analysis of this “dynamically dominant”

subfilter scale content for the key variables will provide knowledge and understanding that can underlie, for example, new advanced modeling strategies.

A. Method to Identify Dynamically Dominant Interactions

In order to arrange and down select the dynamically dominant couplings between SFS and RS motions due to the advective nonlinearity in the LES-filtered governing equations, we take advantage of the elemental representation of inter-scale couplings in multi-dimensional Fourier space and its physical-space counterpart using the “shear wave” concept [23] introduced in Sec. II.

The dynamical evolution of a single RS Fourier mode of a primary variable $\widehat{\beta}(\vec{k}^r)$ occurs within a summation over all Fourier modes $\vec{p}_\Delta$ in the RHS of Eq. (2), which results in a list of triads representing all the inter-scale interactions among Fourier modes $\vec{k}^r$, $\vec{p}_\Delta$, and $\vec{q} = \vec{k}^r - \vec{p}_\Delta$. The same description holds for its physical-space counterpart in Eq. (6), where each triad is a physical-space shear wave $\vec{\Delta}_{sw}(\vec{x}; \vec{k}^r, \vec{p}_\Delta, \vec{q})$ with specific direction, length scale, amplitude, and phase. The sum of all triads associated with a single RS Fourier mode $\vec{k}^r$ recovers the advective nonlinearity $\partial \vec{\beta}_{sw}(\vec{x}; \vec{k}^r)/\partial t$ in the LHS of Eq. (6). To simplify the mathematical notation in this section, we omit the dependence on $\vec{q}$ and indicate each triad within the summation over $\vec{p}_\Delta$ using subscript j , so that $\vec{\Delta}_{sw}(\vec{x}; \vec{k}^r, \vec{p}_\Delta, \vec{q})$ is replaced by $\vec{\Delta}_{sw,j}(\vec{x}; \vec{k}^r)$ and the triads are listed from $j = 1$ to N , where N is the total number of triads.

In order to rank and sort the list of triads of a single RS Fourier mode $\vec{k}^r$, we compare in physical space each triad $\vec{\Delta}_{sw,j}(\vec{x}; \vec{k}^r)$ with $\partial \vec{\beta}_{sw}(\vec{x}; \vec{k}^r)/\partial t = \sum_{j=1:N} \vec{\Delta}_{sw,j}(\vec{x}; \vec{k}^r)$ using a “correlation criterion” defined as

$$\epsilon_j(\vec{k}^r) = \frac{\int_V \vec{\Delta}_{sw,j}(\vec{x}; \vec{k}^r) \cdot \partial \vec{\beta}_{sw}(\vec{x}; \vec{k}^r)/\partial t \, dV}{\int_V \left| \partial \vec{\beta}_{sw}(\vec{x}; \vec{k}^r)/\partial t \right|^2 \, dV} \quad (10)$$

where V is a volume of interest taken as the entire computational domain. The magnitude and sign of $\epsilon_j(\vec{k}^r)$ in Eq. (10) provide information regarding the magnitude and phase correlations, respectively, between each triad $\vec{\Delta}_{sw,j}(\vec{x}; \vec{k}^r)$ and the sum of all triads $\partial \vec{\beta}_{sw}(\vec{x}; \vec{k}^r)/\partial t$. For each $\vec{k}^r$, the list of triads is sorted based on the magnitude $|\epsilon_j|$, so that $|\epsilon_j| \geq |\epsilon_{j+1}|$, and then separated in two lists based on the sign of ϵ_j . The first triads in the sorted list are considered “dynamically dominant” relative to other triads based on the quantification of $|\epsilon_j|$. The sorted triads satisfying the condition $\epsilon_j = \epsilon_j^+ \geq 0$ are grouped in the “positive” set indicated by superscript $+$ (i.e., $\vec{\Delta}_{sw,j}^+(\vec{x}; \vec{k}^r)$ is positive-correlated with $\partial \vec{\beta}_{sw}(\vec{x}; \vec{k}^r)/\partial t$). The remaining sorted triads that satisfy the condition $\epsilon_j = \epsilon_j^- < 0$ are grouped in the “negative” set indicated by superscript $-$ (i.e., $\vec{\Delta}_{sw,j}^-(\vec{x}; \vec{k}^r)$ is negative-correlated with $\partial \vec{\beta}_{sw}(\vec{x}; \vec{k}^r)/\partial t$). As such, the LHS of Eq. (6) can be written in terms of two sorted list of triads

$$\frac{\partial \vec{\beta}_{sw}(\vec{x}; \vec{k}^r)}{\partial t} = \frac{\partial \vec{\beta}_{sw}^+(\vec{x}; \vec{k}^r)}{\partial t} + \frac{\partial \vec{\beta}_{sw}^-(\vec{x}; \vec{k}^r)}{\partial t}, \quad (11)$$

where the sum of positive and negative-correlated triads is $\partial \vec{\beta}_{sw}^{\pm}(\vec{x}; \vec{k}^r)/\partial t = \sum_{j=1:N^{\pm}} \vec{\Delta}_{sw,j}^{\pm}(\vec{x}; \vec{k}^r)$ and $N = N^+ + N^-$. To illustrate the operations described above, Fig. 7a shows the magnitude of ϵ_j^+ and ϵ_j^- for a single RS Fourier mode $\vec{k}^r$ from the work summarized in Sec. IV, where we have approximately $N^+ \approx N^- \approx 3 \times 10^4$ triads in each sorted list.

In order to down select the dynamically dominant triads within each set, a “truncation criterion” was defined such that the partial sum of the truncated set of triads recovers a specified (large) percentage of the total sum. The truncation criterion for the positive and negative set of triads is given as

$$\zeta_m^{\pm}(\vec{k}^r) = \frac{\int_V \left[\sum_{j=1:m} \vec{\Delta}_{sw,j}^{\pm}(\vec{x}; \vec{k}^r) \right] \cdot \partial \vec{\beta}_{sw}^{\pm}(\vec{x}; \vec{k}^r)/\partial t \, dV}{\int_V \left| \partial \vec{\beta}_{sw}^{\pm}(\vec{x}; \vec{k}^r)/\partial t \right|^2 \, dV} \quad (12)$$

for $m = 1 : N^{\pm}$. The criterion in Eq. (12) represents the normalized correlation between the sum of the truncated set of triads $\sum_{j=1:m} \vec{\Delta}_{sw,j}^{\pm}(\vec{x}; \vec{k}^r)$ and the sum of all triads in the set $\partial \vec{\beta}_{sw}^{\pm}(\vec{x}; \vec{k}^r)/\partial t$ for the positive and negative sets. By

construction the triads within each set have the same phase-correlation (i.e, same sign of ϵ_j) and are sorted based on correlation magnitude (i.e, $|\epsilon_j|$), so that $\zeta_m^{+,-}(\vec{k}^r)$ is a smooth monotonically increasing function where $0 < \zeta_m^{+,-}(\vec{k}^r) \leq 1$ and $\zeta_{N^{+,-}}^{+,-}(\vec{k}^r) = 1$. For a given $\vec{k}^r$, each set is then truncated based on user-defined truncation criterion ζ^* , such that $\zeta_{N_{subset}^{+,-}}^{+,-} = \zeta^*$, producing two subsets containing significantly smaller number of triads $N_{subset}^{+,-} \ll N^{+,-}$. The final set of dynamically dominant triads is obtained by regrouping the triads associated with the truncated positive and negative sets, such that

$$\frac{\partial \vec{\beta}_{sw}(\vec{x})}{\partial t} = \sum_{j=1:N} \vec{\Delta}_{sw,j}(\vec{x}) \approx \sum_{j=1:N_{subset}^+} \vec{\Delta}_{sw,j}^+(\vec{x}) + \sum_{j=1:N_{subset}^-} \vec{\Delta}_{sw,j}^-(\vec{x}). \quad (13)$$

Figure 7b shows the truncation criterion ζ_m^+ and ζ_m^- for the same RS Fourier mode $\vec{k}^r$ of Fig. 7a, where the vertical dashed lines indicate the truncated number of triads in the positive $N_{subset}^+ \approx 400$ and negative $N_{subset}^- \approx 80$ subsets using $\zeta^* = 0.85$, which is significantly smaller than the total number of triads in each set.

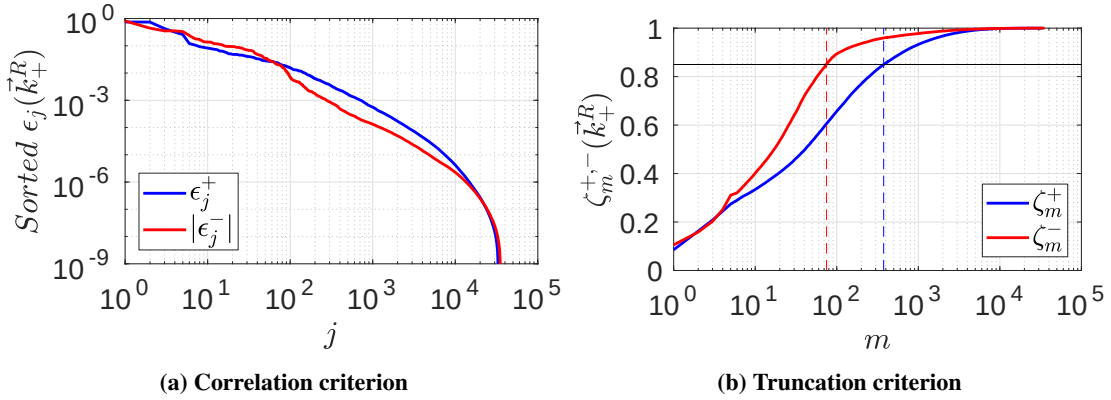


Fig. 7 (a) Correlation and (b) truncation criterion for triads associated with a single RS Fourier mode $\vec{k}^r$ for $\vec{\beta} = \rho \vec{u}$ from the work summarized in Sec. IV.

The procedure described above is applied to all individual Fourier modes within the resolved-scales $\vec{k}^r > 0$ using a truncation criterion $\zeta^* = 85\%$ to the three primary variables of interest in this study ($\vec{\beta} = \rho \vec{u}$, ρh , and ρY_{HCO}). For each individual RS Fourier mode $\vec{k}^r$, we extract the participating wave-vectors $\vec{p}_\Delta$ and $\vec{q}$ within the SFS region from the truncated list of dynamically dominant triads. The collection of all SFS wave-vectors that are dynamically dominant for all individual RS Fourier modes $\vec{k}^r$ for a given primary variable $\vec{\beta}$ defines the dynamically dominant SFS space, which is identified by superscript ds . The associated dynamically dominant SFS content is given by the collection of Fourier modes $\hat{\vec{\beta}}(\vec{p}_\Delta^{ds})$ and $\hat{\vec{u}}(\vec{q}_\beta^{ds})$, where subscript β indicate the dynamically dominant SFS modes of the velocity field associated with the primary variable $\vec{\beta}$. The inverse Fourier transform of this collection of Fourier coefficients results in the physical-space counterpart of the dynamically dominant SFS content given by $\vec{\beta}^{ds}(\vec{x})$ and $\vec{u}_\beta^{ds}(\vec{x})$. The LHS of Eq. (1) can be recomputed using the dynamically dominant SFS content as,

$$(\partial \vec{\beta}^r / \partial t)_{DS} = -\nabla \cdot \left(\vec{\beta}^r \vec{u}^r + \vec{\beta}^r \vec{u}_\beta^{ds} + \vec{\beta}^{ds} \vec{u}^r + \vec{\beta}^{ds} \vec{u}_\beta^{ds} \right)^r, \quad (14)$$

where subscript DS implies that only the dynamically dominant SFS interacts with the resolved scales.

The choice of the truncation criterion ζ^* provides a reduction in the number of dynamically dominant SFS wave-vectors $\vec{p}_\Delta^{ds}$ and $\vec{q}_\beta^{ds}$ for a given $\vec{k}^r$ mode. As the procedure is applied to all individual $\vec{k}^r$, we identify a collection of SFS wave-vectors that is dynamically dominant considering triads for all $\vec{k}^r$. However, for a single $\vec{k}^r$, this collection includes non-dominant SFS wave-vectors that are dynamically dominant for other $\vec{k}^r$ modes, so that a larger percentage of the advective nonlinearity is effectively recovered for this single $\vec{k}^r$. In order to check the extent that the energy content in the advective nonlinearity for the evolution of the RS motion (i.e., all individual $\vec{k}^r$) is recovered with the

reduced subset of dynamically dominant SFS modes (i.e., all $\vec{p}_\Delta^{ds}$ and $\vec{q}_\beta^{ds}$), we compute

$$\alpha_{\vec{\beta}}(\zeta^*) = \frac{\int_V \left| (\partial \vec{\beta}^r / \partial t)_{DS} - \partial \vec{\beta}^r / \partial t \right|^2 dV}{\int_V \left| \partial \vec{\beta}^r / \partial t \right|^2 dV}, \quad (15)$$

for $\vec{\beta} = \rho \vec{u}$, ρh , and ρY_{HCO} . Figures 8a-b show the dependence of $\alpha_{\vec{\beta}}(\zeta^*)$ (Eq. (15)) and the number of dynamically dominant SFS modes relative to all SFS modes within the SFS flame-scale domain $|\vec{k}| \delta_f / \pi \leq 1$ ($n_{DS,f} / n_{S,f}$) to the choice of user-defined truncation criterion ζ^* , where the dashed vertical line indicates $\zeta^* = 85\%$ that makes Eq. (15) smaller than 1% for all variables. Figure 8 indicates that the energy content of the advective nonlinearity in the LHS of Eq. (1) is preserved although the procedure described above provides a dramatic reduction in the number of SFS modes dynamically dominant for the evolution of the resolved scales due to the advective nonlinearity.

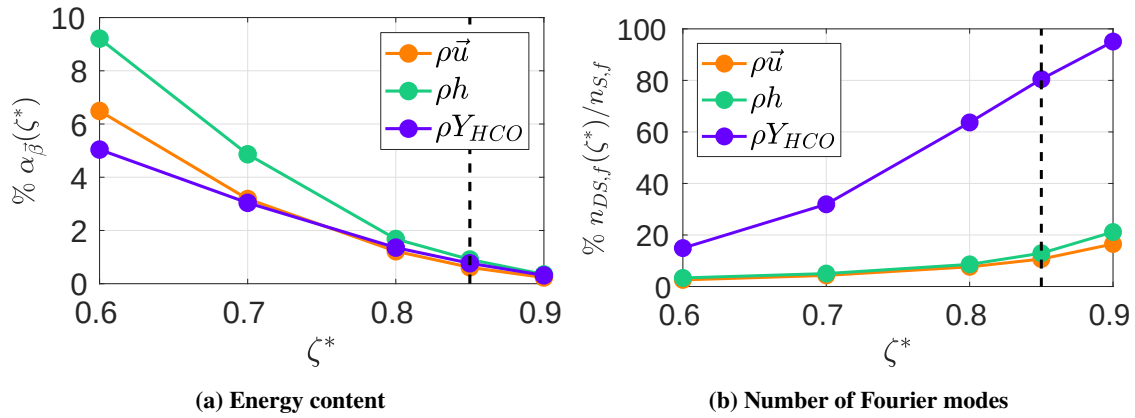


Fig. 8 (a) Deviation in energy content of the advective nonlinearity reconstructed with dynamically dominant SFS content and (b) number of dynamically dominant SFS modes within the flame-scale domain ($|\vec{k}| \delta_f / \pi \leq 1$) as a function of user-defined truncation criterion ζ^* .

B. Dynamically Dominant Sub-Filter Scale Content: Fourier Space

The dynamically dominant SFS content in Fourier space of each primary variable $\vec{\beta}(\vec{p}_\Delta^{ds})$ ($\vec{\beta} = \rho \vec{u}$, ρh , and ρY_{HCO}) is shown in Fig. 9. Consistent with Fig. 6, we plot iso-contours of the logarithmic squared Fourier coefficients for each primary variable, but only the dynamically dominant SFS modes are colored, so that resolved scales or nondominant SFS are indicated by the white iso-level. We make use of the symmetries discussed in Sec. IV.C to show the squared Fourier coefficients of the streamwise and cross-stream components of the momentum vector in the top and bottom halves of Fig. 9a.

Figure 9 shows that the dynamically dominant SFS modes selected by the method represent only a very small percentage of all SFS Fourier modes for $\rho \vec{u}$ and ρh . However, this is not the case for ρY_{HCO} . In fact, whereas only 13% of all SFS modes within the flame-scale spectral domain are dynamically important for $\rho \vec{u}$ and ρh , 80% are important for ρY_{HCO} (as observed in Fig. 8b for $\zeta^* = 0.85$). Much of the dynamically important SFS content of ρY_{HCO} extends outside the flame-scale domain. For all variables analyzed, the dynamically dominant SFS spectral modes extend from the SFS boarder with the RS cutoff wavenumbers into the higher wavenumbers (small scales) following the star-like pattern described in Sec. IV. For ρu , there is a clear correlation between the location of the dynamically dominant SFS modes (upper half of Fig. 9a) and the more energetic Fourier modes within the star-like pattern (upper half of Fig. 6a). This correlation is less clear for ρh and ρY_{HCO} indicating that the dynamically dominant Fourier content includes both the legs of the star-like pattern and spectral regions spread over a broader range of angles. The dynamically dominant SFS modes for velocity components are observed (not shown here) to be approximately the same as the dynamically dominant SFS modes for the respective primary variables (i.e., collection of wave-vectors $\vec{p}_\Delta^{ds}$ is approximately the same

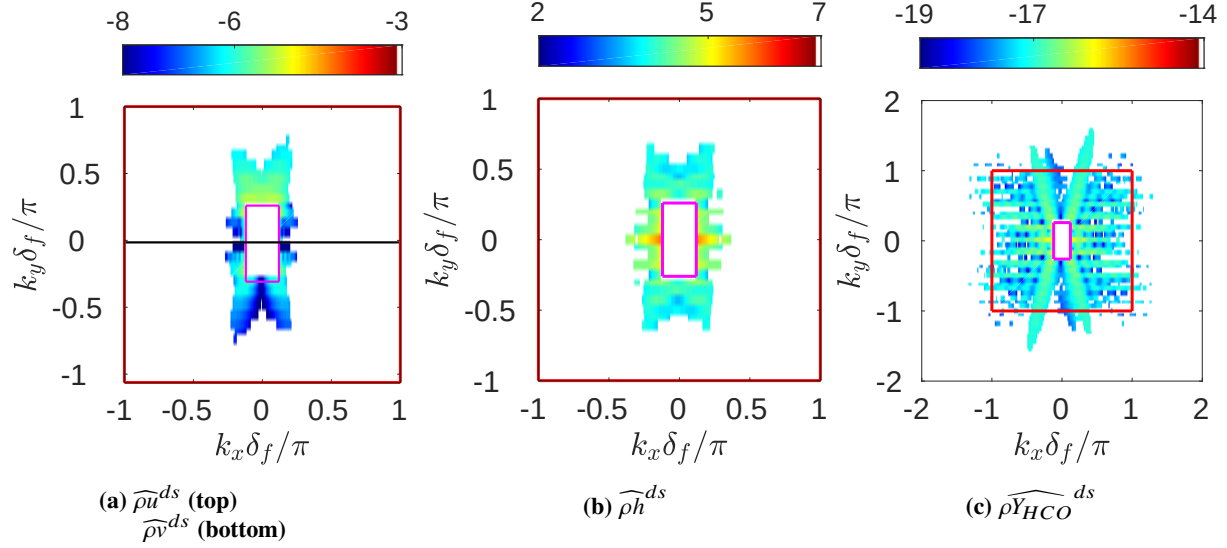


Fig. 9 Iso-contours of the logarithmic squared Fourier coefficients of dynamically dominant SFS modes for the primary variables shown in Fig. 6. White iso-level indicates Fourier modes at resolved scales or nondominant SFS. (a) $\log_{10}(\widehat{\rho u}^{ds})$ (top) and $\log_{10}(\widehat{\rho v}^{ds})$ (bottom); (b) $\log_{10}(\widehat{\rho h}^{ds})$; (c) $\log_{10}(\widehat{\rho Y_{HCO}}^{ds})$.

of $\vec{q}_{\vec{\beta}}^{ds}$), so that dominant SFS modes are contained within the flame-scale domain for momentum and enthalpy and extend outside for formyl radical concentration.

In order to better understand the distinct collections of dynamically dominant SFS modes for each primary variable, we recall from Eq. (2) that the global characteristics of each triad depends on its geometry (i.e., wave-vectors), relative orientation between $\vec{k}^r$ and $\vec{u}(\vec{q})$, magnitude and the phase relationships between $\vec{\beta}(\vec{p}_{\Delta})$ and $i\vec{k}^r \cdot \vec{u}(\vec{q})$ [23]. As described in Sec. IV, the SFS Fourier content for all primary variables is organized following a star-like pattern, with a faster decay from large to small scales for ρu , ρv , and ρh , but slower decay for ρY_{HCO} . As a result of the different decay of Fourier content over the star-like pattern for distinct primary variables, the dynamically important SFS modes for momentum and enthalpy do not extend beyond $|\vec{k}|\delta_f/\pi \approx 0.8$, while for ρY_{HCO} goes towards $|\vec{k}|\delta_f/\pi \approx 1.5$.

The extent that the dynamically dominant SFS modes in ρY_{HCO} extend much farther away in spectral space from the RS cutoff wavenumbers reflects the dominant SFS dynamics associated with the generation of thermal energy (i.e., heat release) within the reaction zone. As per discussion of Sec. I, key dynamics underlying the function of combustion devices are almost always dominantly in the unresolvable SFS motions, as now evident from the dynamically dominant SFS content of ρY_{HCO} (Fig. 9c) and its associated velocity field (not shown here).

C. Dynamically Dominant Sub-Filter Scale Content: Physical Space

Figure 10 shows the physical-space counterpart of the dynamically dominant SFS content for each primary variable $\vec{\beta}^{ds}(\vec{x})$ ($\vec{\beta} = \rho \vec{u}$, ρh , and ρY_{HCO}), where bottom and top halves of each panel present entire SFS signal and dynamically dominant SFS signal, respectively. The two-dimensional mean value of the respective original signals is added for a direct comparison with Fig. 5 and the two dotted curves indicate the RS flame front defined by RS progress variable iso-levels of 5% and 85%. The “ghost ripples” observed in Fig. 10 are a result of the sharp filters (i.e., inverse Fourier transform of specific SFS Fourier modes) employed to obtain the physical-space signals, referred as “ringing effect”. We observed (not shown here) that this kinematic ringing effect can be removed with the use of smooth Fourier space filters as representative of true smooth filters in operational LES. We find that appropriate smooth filters remove the ghost ripples away from the RS flame front without significant modification to the signal over regions with large magnitude within the RS flame front.

Figure 10 shows that the entire SFS content (bottom halves of each iso-contour) of the three primary variables analyzed in this work is localized within the RS flame front. Although the Fourier basis is non-local in physical space (Eq. (5)), the phase relationships within collections of SFS modes is shown to localize the physical-space content in the regions with largest gradients over the reaction zone within the flame front in Fig. 10. Yet, significant structural

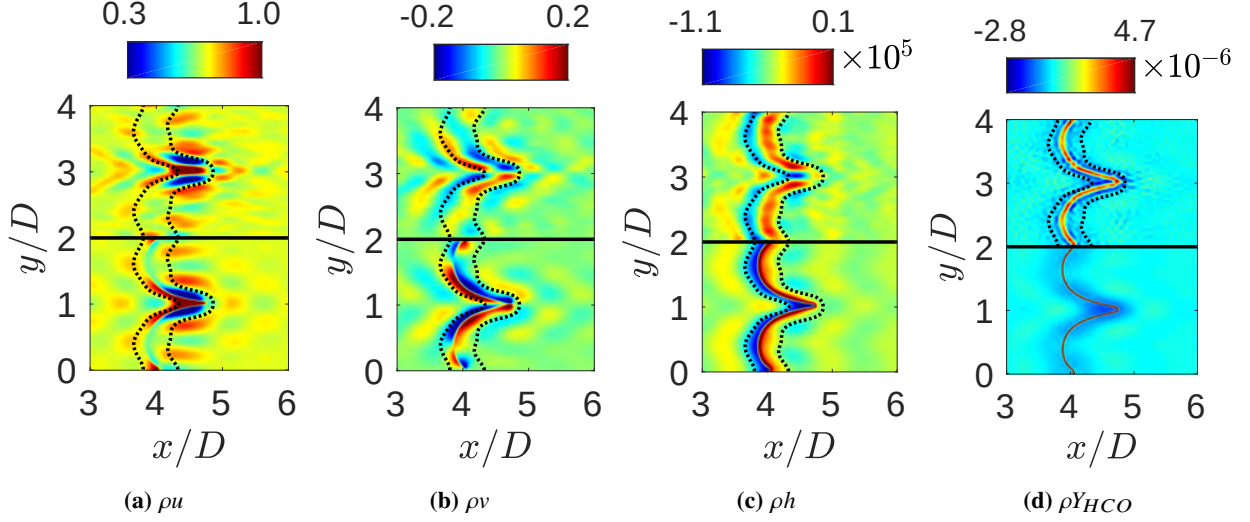


Fig. 10 Iso-contours of (a-b) streamwise and cross-stream components of momentum (ρu and ρv in $\text{kg/m}^2\text{s}$) vectors, (c) mixture enthalpy (ρh in J/m^3), and (d) formyl radical concentration (ρY_{HCO} in kg/m^3), respectively. Entire SFS and dynamically dominant SFS signals are shown in the bottom and top half of each panel, respectively.

differences in the SFS content are observed for distinct variables. The SFS content of ρu and ρv represents the small-scale content of the flame cusps, while ρh represents the streamwise and corrugated flame fronts resulting in the double-banded structure in physical space surrounding the reaction zone. In contrast, ρY_{HCO} shows a much thinner high-gradient structure that peaks in the reaction zone within the RS flame front. The truncation of dynamically dominant SFS modes removes small-scale features in the SFS signals of all three primary variables, without fundamental modification in the structure of the SFS signal described above. This is observed in the top halves of the domain in Fig. 10, where the smallest-scale gradients within the reaction zone are removed from ρu^{ds} , ρv^{ds} , and ρh^{ds} , while the thin regions where ρY_{HCO}^{ds} peaks become thicker in ρY_{HCO}^{ds} .

For primary variables that have most of its content at large scales and fast decay of energy towards small scales (i.e., $\rho \vec{u}$ and ρh), the large-scale features of the dynamically dominant SFS content are shown to be coupled with the smallest resolved-scales representing the corrugations and thickness of the RS flame front. This indicates that energetic features of the dynamically dominant SFS content are directly coupled with characteristics at the resolved scales. In contrast, the dynamically dominant SFS content of ρY_{HCO} is shown to follow the smallest corrugations of the flame front centerline, which deviate from the RS flame centerline in regions with higher corrugations, such as the flame cusps. These results are potentially valuable in the design of new SFS modeling strategies for key variables in LES of premixed turbulent combustion wherein only RS data on the LES grid are available, in principle.

VI. Conclusions

The present work addresses Stage 3 of the research strategy illustrated in Fig. 1, the development of a procedure to identify dynamically dominant inter-scale couplings and isolate the dynamically dominant SFS content within these couplings for application to LES of premixed turbulent combustion. Two other studies as part of this strategy are described in [21, 22]. Here we make use of the previous analyses from Stages 1 and 2 [21, 22] that relate spectral content to physical space content of key variables in premixed turbulent combustion (summarized in Sec. IV) to characterize nonlinear triadic inter-scale couplings between RS and SFS motions. We take advantage of the mathematical simplicity of the Fourier description of the governing equations, where advective nonlinearities appear with a “triadic form” (Sec. II), to develop a procedure to arrange and down-select the dynamically dominant triadic couplings between SFS and RS motions (Sec. V.A). From these groups of dominant RS-SFS couplings, we determined the SFS content that is dynamically coupled to the advective terms that contribute to the nonlinear evolution of RS momentum, enthalpy, and key species. The procedure is applied to the three primary variables analyzed in this work ($\vec{\beta} = \rho \vec{u}$, ρh , and ρY_{HCO}).

The dynamically dominant SFS modes in the variables of interest $\hat{\vec{\beta}}(\vec{p}_{\Delta}^{ds})$ are shown (Sec. V.B) to represent a very

small fraction of all SFS modes in the dynamical system. Whereas only 13% of all SFS modes within the flame-scale spectral domain are dynamically important for ρu and ρh , 80% are important for ρY_{HCO} . The much larger percentage of dynamically dominant SFS modes for ρY_{HCO} is shown to be associated with the observation that the variances of key species concentrations underlying heat release are largely unresolvable by a LES grid designed to capture a large percentage of variance in momentum and enthalpy. This SFS content is observed to lie largely along “star-like patterns” in 2D Fourier space associated with small-scale content along the flame front. Compared to other variables, this key intermediate specie displays a much slower decay of Fourier content along the star-like pattern. Thus, the RS-SFS couplings in ρY_{HCO} become as dynamical significant as the fully resolved inter-scale couplings in the LES equations. The corresponding SFS modeled terms for ρY_{HCO} therefore require more accurate SFS representation from both structural and energetic perspectives in the LES framework than do the modeled advective terms for momentum and enthalpy, which typically apply an eddy diffusivity representation.

The physical-space counterpart of the dynamically dominant SFS Fourier content for each primary variable is shown (Sec. V.C) to capture the overall structure of the SFS signals without the smallest-scale gradients within the flame cusps and the reaction zone. More interestingly, the larger-scale SFS features of the dynamically dominant SFS content for ρu and ρh are shown to be coupled with the smallest resolved motions that characterize the corrugations and thickness of the resolved flame front. This indicates that some key structural features at SFS are coupled with RS structures, which may be useful to capture with advanced LES modeling strategies. This is not the case for key combustion intermediates such as ρY_{HCO} , where the dynamically dominant SFS content is shown to follow the smallest-scale corrugations of the flame front centerline.

The research project illustrated in Fig. 1 provides evidence of particular difficulties in LES of turbulent combustion systems associated with chemical kinetics that produce key dynamics outside the essential LES framework. In future research, the tools developed in this work will be applied to three-dimensional reduced-physics simulations consisting of interactions between vortex rings and a premixed flame, and then to fully turbulent premixed flame-turbulence interactions.

Acknowledgments

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A. Removing Pollution from Boundary Discontinuities in Non-Periodic Direction of Signals over Finite Domains: A “Discontinuity Pollution Removal” Procedure

The discontinuity pollution removal procedure developed in [21] is summarized here considering a smooth and continuous 2D signal $f(0 \leq x \leq L_x, 0 \leq y \leq L_y)$ defined over a finite domain non-periodic and periodic in x and y directions, respectively, so that the boundary discontinuity is defined as, $\Delta f(y) = f(L_x, y) - f(0, y)$. The Fourier transform of $f(x, y)$ results in Fourier coefficients $\widehat{f}(k_x, k_y)$, where k_x and k_y are the discrete wavenumbers. The fundamental C^0 boundary discontinuity function in physical and Fourier space is given by the Fourier pair,

$$\widehat{d}(k_x, k_y) = \frac{i}{k_x} \frac{\Delta \widehat{f}(k_y)}{L_x} \leftrightarrow d(x, y) = \frac{\Delta f(y)}{L_x} \left(x - \frac{L_x}{2} \right), \quad (\text{A.1})$$

for $k_x \neq 0$ and $\vec{x}$ within the finite domain. In physical space, the boundary discontinuity is given by the saw-tooth function in x direction with variations in y controlled by $\Delta f(y)$, while its Fourier-space counterpart consists of power-law functions in k_x direction with variations in k_y controlled by $\Delta \widehat{f}(k_y)$.

The discontinuity pollution removal procedure consists of removing at each k_y wavenumber the power-law pollution towards large k_x from the original signal using a high-pass Fourier space filter in k_x to modulate $\widehat{d}(k_x, k_y)$. The modulation function is constructed as,

$$m(k_x; k_{x, \text{cut}}(k_y)) = \begin{cases} 1 & \text{if } |k_x| \geq k_{x, \text{cut}}(k_y) \\ 0 & \text{if } |k_x| < k_{x, \text{cut}}(k_y) \end{cases}, \quad (\text{A.2})$$

and the modulated power-law in Fourier space is,

$$\widehat{d}_m(k_x, k_y; k_{x,cut}(k_y)) = m(k_x; k_{x,cut}(k_y)) \widehat{d}(k_x, k_y), \quad (\text{A.3})$$

where its physical-space counterpart $d_m(x, y; k_{x,cut}(k_y))$ is obtained from its inverse transform. As a result, $d_m(x, y; k_{x,cut}(k_y))$ is given by harmonic functions in x direction with smooth decay towards zero in the interior of the domain and variations in y controlled by $\Delta f(y)$. The final physical-space signal is obtained by removing the modulated discontinuity from the original signal,

$$f_{d_m}(x, y; k_{cut}(k_y)) = f(x, y) - d_m(x, y; k_{cut}(k_y)). \quad (\text{A.4})$$

To identify the optimal filter cutoff wavenumber $k_{x,cut}^{opt}(k_y)$ that provides the best tradeoff between the signal modification and the residual boundary discontinuity, a quasi-1D analysis at each k_y wavenumbers is employed. For that, the Fourier coefficients of $\widehat{f}(k_x, k_y)$ are filtered at fixed wavenumbers $|k_y| = k_y^+$, where k_y^+ is the positive side of the wavenumber coordinate k_y . Fourier modes at these two wavenumbers ($\pm k_y^+$) are symmetric with respect to the wavenumber origin since for real signals the “reality condition” states that, $\widehat{f}(-\vec{k}) = \widehat{f}^*(\vec{k})$. The filtered signal is defined as,

$$\tilde{f}(x, y) = IFFT \left[\delta(|k_y| - k_y^+) \widehat{f}(k_x, k_y) \right], \quad (\text{A.5})$$

where $\delta(|k_y| - k_y^+)$ is a Dirac delta function used to filter the signal at k_y indicated by superscript $\sim$.

The nondimensional signal modification parameter at each k_y^+ wavenumber is defined as,

$$\epsilon(k_y^+; k_{x,cut}(k_y^+)) = \frac{\|\tilde{d}_m(x, y; k_{x,cut}(k_y^+))\|_V}{\|\tilde{f}(x, y)\|_V}. \quad (\text{A.6})$$

where $\|\tilde{f}(x, y)\|_V$ indicates the L^2 norm of the filtered signal $\tilde{f}(x, y)$ within the volume of interest V computed after removing the its mean value. Similarly, the nondimensional residual jump parameter is defined as,

$$\Delta_r(k_y^+; k_{x,cut}(k_y^+)) = \max_{(y)} \left\{ \frac{|\Delta \tilde{f}_{d_m}(y; k_{x,cut}(k_y^+))| / \Delta x}{|\max_{(x)} (\partial \tilde{f} / \partial x) (y)|} \right\} \quad (\text{A.7})$$

where $\Delta \tilde{f}_{d_m} = \tilde{f} - \tilde{f}_{d_m}$ is the jump of $\tilde{f}_{d_m}$ over the x direction and $\max_{(y)}$ indicates the maximum values in y direction.

To identify the optimal filter cutoff wavenumber $k_{x,cut}^{opt}(k_y^+)$ at each k_y^+ wavenumbers a weighted criteria is constructed as,

$$\phi(k_{x,cut}(k_y^+); \alpha) = \alpha \epsilon(k_y^+; k_{x,cut}(k_y^+)) + (1 - \alpha) \Delta_r(k_y^+; k_{x,cut}(k_y^+)) \quad (\text{A.8})$$

where $\alpha = 0.85$ is used to weight signal modification and residual boundary discontinuity for this work. The optimal filter cutoff wavenumber is found minimizing $\phi(k_{x,cut}(k_y^+); \alpha)$ in Eq. (A.8) at each k_y^+ ,

$$\phi(k_{x,cut}^{opt}(k_y^+); \alpha) = \min [\phi(k_{x,cut}(k_y^+); \alpha)]. \quad (\text{A.9})$$

The procedure is performed at each k_y^+ so that the optimal boundary discontinuity function $d_m(x, y; k_{cut}^{opt}(k_y))$ can be computed and removed from the original signal.

B. “Reality Condition” for Triad Pairs

As per discussion of Sec. II, the “reality condition” creates symmetry in Fourier space with respect to the wavenumber origin $\widehat{\phi}(-\vec{k}^r) = \widehat{\phi}^*(\vec{k}^r)$, so that the wave-vector space is separable into two subsets and the projection of the Fourier coefficient pairs may be rewritten over the positive set $\vec{k}_+^r$ using Eq. (5). The same reality condition can be found for pairs of triads within the summation in the RHS of Eq. (3) as,

$$\begin{aligned} \widehat{\Delta}(-\vec{k}_+^r, \vec{p}_\Delta; \vec{q}) &= \widehat{\beta}(\vec{p}_\Delta) \left(-i\vec{k}_+^r \right) \cdot \widehat{u}(\vec{q}) = \widehat{\beta}^*(-\vec{p}_\Delta) \left(i\vec{k}_+^r \right)^* \cdot \widehat{u}^*(-\vec{q}) = \dots \\ &\dots \left\{ \widehat{\beta}(-\vec{p}_\Delta) \left(i\vec{k}_+^r \right) \cdot \widehat{u}(-\vec{q}) \right\}^* = \widehat{\Delta}^*(\vec{k}_+^r, -\vec{p}_\Delta, -\vec{q}). \end{aligned} \quad (\text{B.1})$$

Thus, the RHS of Eq. (7) can be obtained as,

$$\begin{aligned}
 \left. \frac{\partial \vec{\beta}^r(\vec{x})}{\partial t} \right|_{adv} &= \sum_{\vec{k}^r} \sum_{\vec{p}_\Delta} \widehat{\Delta}(\vec{k}^r, \vec{p}_\Delta; \vec{q}) e^{i\vec{k}^r \cdot \vec{x}} = \dots \\
 &\dots \sum_{\vec{k}_+^r} \left\{ \sum_{\vec{p}_\Delta} \widehat{\Delta}(\vec{k}_+^r, \vec{p}_\Delta; \vec{q}) e^{i\vec{k}_+^r \cdot \vec{x}} + \sum_{\vec{p}_\Delta} \widehat{\Delta}(-\vec{k}_+^r, \vec{p}_\Delta; \vec{q}) e^{-i\vec{k}_+^r \cdot \vec{x}} \right\} = \dots \\
 &\dots \sum_{\vec{k}_+^r} \sum_{\vec{p}_\Delta} \left\{ \widehat{\Delta}(\vec{k}_+^r, \vec{p}_\Delta; \vec{q}) e^{i\vec{k}_+^r \cdot \vec{x}} + \widehat{\Delta}^*(\vec{k}_+^r, -\vec{p}_\Delta, -\vec{q}) e^{-i\vec{k}_+^r \cdot \vec{x}} \right\} = \sum_{\vec{k}_+^r} \sum_{\vec{p}_\Delta} \vec{\Delta}_{sw}(\vec{x}; \vec{k}_+^r, \vec{p}_\Delta, \vec{q}). \quad (\text{B.2})
 \end{aligned}$$

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Appendix C

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Dynamically Dominant Interscale Couplings in the Nonlinear Chemical Source Terms for Species Evolution in Premixed Turbulent Combustion with Application to LES Modeling

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Abstract: In large-eddy simulation (LES) of premixed turbulent combustion, subfilter scale (SFS) motions directly impact the dynamical evolution of resolved scale (RS) motions through the nonlinear interscale interactions within the advective and chemical source terms. Using simulations of the interactions between trains of vortices and an initially planar laminar flame to model local turbulence-flame dynamics, we quantify the dominant nonlinear couplings underlying the evolution of species concentration for application to SFS closure models in the LES framework. Applying a new Fourier-space-based methodology applicable to inhomogeneous turbulence, we quantify the SFS content that dominantly contributes to the resolved-scale evolution of key chemical species associated with second-order chemical reactions. We take advantage of the geometrical simplicity of the Fourier spectral description of the governing equations where third-order nonlinearities are given by a sum of elemental quadratic interactions involving four Fourier modes whose wave-vectors form a quadrangle in multidimensional Fourier space (“quadrads”). By reorganizing this elemental summation, we down-select the nonlinear RS-SFS quadrads that dominantly contribute to the time derivative of RS species concentration. From these dominant quadrads we extract the corresponding “dominant” SFS content and encapsulate this content within physical-space representations for potential application to advanced SFS modeling strategies.

Keywords: *Chemical reaction, Premixed turbulent combustion, Large-eddy simulation, SGS modeling*

1. The Large-Eddy Simulation Application in Premixed Turbulent Combustion

Computational Fluid Dynamics (CFD) has been used as an important tool for design of next generation advanced combustion devices for aerospace applications [1]. With ever-increasing power and advances in computing technologies, Large-Eddy Simulations (LES) is becoming a feasible advanced tool for use in the design of practical aerospace systems and devices [2]. In the classical LES framework, the numerical solution of the dynamical equations must be carried out on a grid that resolves well a large percentage of the variances of the most energetic scales of motion under the assumption that those turbulence variables are dominated by the forward cascade of momentum and energy from larger to smaller turbulence length scales [3]. If this is the case, the evolution of

the more energetic grid-filtered motions are dominated by the dynamical interactions explicitly resolved on the grid, and the dynamical effects of the unresolved Sub-Filter-Scale (SFS) motions on the evolution of the Resolved-Scale (RS) motions are higher order. When this is the case, first and second order statistics are accurately predicted in LES. (In contrast, Reynolds-averaged Navier-Stokes simulations are characterized by the need to model second order statistics at the first order level).

However, the predictive capabilities of LES for premixed turbulent combustion systems are limited by a fundamental deviation from the classical LES framework. In these systems, the chemical reactions dominantly associated with the chemical generation of thermal energy (i.e., heat release), generally occurs at scales unresolvable by the LES grid [4, 5]. Consequently, some key dynamics underlying the function of combustion devices are almost always dominantly in the unresolved SFS motions so that basic assumptions that make LES an accurate predictive tool break down.

In previous work [6], we analyzed kinematic relationships between Fourier-physical space coherent structure in key dynamic variables in premixed flame-turbulence interaction in context with the LES decomposition into grid-resolved and subfilter scales. In [7], we quantified dynamically dominant advective inter-scale couplings between RS and SFS necessary for more accurate prediction of the evolution of RS premixed turbulent combustion. The current study addresses the identification of dynamically dominant chemical inter-scale couplings and analysis of the dominant SFS content within these chemical couplings for application to LES of premixed turbulent combustion. The specific objectives are threefold. Firstly, we use the previous framework developed in [6], to relate spectral content to physical space content of key variables in premixed turbulent combustion. Secondly, we use this knowledge to characterize nonlinear chemical inter-scale couplings between RS and SFS as done in [7]. Finally, we identify dynamically dominant chemical inter-scale couplings and extract the SFS content within these couplings.

This manuscript is organized as follows. The general characteristics and mathematical framework of inter-scale couplings in physical and Fourier space are presented in Sec. 2. The design of the reduced-physics simulations consisting of flame-vortex interactions is discussed in Sec. 3. The Fourier-physical space relationships of chemical species are presented in Sec. 4. Sec. 5 presents the overall characteristics, classification, and identification of key chemical reactions within the chemical kinetic model. The chemical inter-scale couplings associated with key reactions are investigated in Sec. 6 and their dynamically dominant content is isolated and analyzed in Fourier and physical space. The key findings of this study are summarized in Sec. 7.

2. Characteristics of Key Chemical Nonlinearities in Physical and Fourier Space

As an inherently nonlinear high degree-of-freedom dynamical system, the interactions among scales of motion in premixed turbulent combustion are central both to the dynamics and simulation/modeling of flame-turbulence interactions. In this work we focus on the dynamical interactions among turbulence scales through the chemical nonlinearities in the filtered governing equations within LES frameworks. It is only through nonlinearities in the filtered governing equations that subfilter scale dynamics can affect resolved scale evolution, which is often referred as ‘closure problem’ in turbulence. The finite-rate chemistry source terms in the RHS of each species transport equation comprises a sum of the contributions from all elementary chemical reactions, j , resulting in the formation and destruction of a given species. Whereas the number of chemi-

cal reactions (j) can be very large with many reactions of interest dominated by collisions of two species (m, n) resulting in second-order reactions [8]. The chemical source term $\dot{\omega}_{i,j}$ for production of species mass concentration ρY_i (mass per unit volume) associated with a second-order reaction j between species Y_m and Y_n may be written as,

$$\dot{\omega}_{i,j} = W_i \kappa_j(T) \left(\frac{\rho Y_m}{W_m} \right) \left(\frac{\rho Y_n}{W_n} \right) \quad (1)$$

where $\kappa_j(T)$ is the temperature-dependent reaction rate function of reaction j (in $m^3/kmol\ s$) and the unity stoichiometric coefficients are omitted for simplicity.

The structure of the chemical nonlinearity associated with the second-order reaction given by Eq. 1 for the evolution of the resolved scale species mass fraction can be written as follows:

$$\left. \frac{\partial \theta_i^r}{\partial t} \right|_{chem} = \dot{\omega}_{i,j}^r = (K_j^r \theta_m^r \theta_n^r)^r + \left(\begin{matrix} K_j^s \theta_m^r \theta_n^r \\ + K_j^r \theta_m^s \theta_n^r \\ + K_j^s \theta_m^s \theta_n^s \end{matrix} \right)^r + \left(\begin{matrix} K_j^r \theta_m^s \theta_n^s \\ + K_j^s \theta_m^r \theta_n^s \\ + K_j^s \theta_m^s \theta_n^r \end{matrix} \right)^r + (K_j^s \theta_m^s \theta_n^s)^r, \quad (2)$$

where we define, $K_j(T) \equiv \frac{W_i \kappa_j(T)}{W_n W_m}$, and, $\theta_n \equiv \rho Y_n$, as the modified temperature-dependent reaction rate function and species mass concentration per unit volume, respectively. The RHS of Eq. 2 represents all inter-scale nonlinear chemical coupling between K_j , θ_m , and θ_n that contribute to the temporal evolution of resolved scale θ_i^r . This third-order nonlinearity results in 8 terms involving either three RS, two RS and one SFS, one RS and two SFS, or three SFS contributions from K_j , θ_m , and θ_n . The first term in the RHS of Eq. 2 represents the chemical nonlinearity fully-resolved on the LES grid referred as “resolved-scale-resolved-scale” (RS-RS) chemical couplings. This should be the leading-order term in the equation of motion for $\vec{\beta}^R$ if the grid is properly designed under the fundamental LES assumptions discussed in Sec. 1. The other terms in the RHS of Eq. 2 describe the “resolved-scale-subfilter-scale” (RS-SFS) couplings that should be higher order in the RHS of Eq. 2 and must be appropriately modeled in LES frameworks.

Fourier-spectral space is employed to explicitly represent the nonlinear couplings between scales through the chemical nonlinearities in Eq. 2, where “scales of motion” are defined with a complex (exponential) basis of elemental spectral content, each of which represents precisely-defined scale, direction, and phase. The structure of the chemical nonlinearities associated with second-order reactions for the evolution of the resolved scales in Fourier space is given by the Fourier transform of Eq. 2 as

$$\left. \frac{\partial \hat{\theta}_i(\vec{k}^r)}{\partial t} \right|_{chem} = \hat{\omega}_i(\vec{k}^r) = \sum_{\vec{p}_{\square}} \sum_{\vec{q}_{\square}} \widehat{K}_j(\vec{p}_{\square}) \widehat{\theta}_m(\vec{q}_{\square}) \widehat{\theta}_n(\vec{l}) = \sum_{\vec{p}_{\square}} \sum_{\vec{q}_{\square}} \widehat{\square}(\vec{k}^r, \vec{p}_{\square}, \vec{q}_{\square}; \vec{l}), \quad (3)$$

where $i = \sqrt{-1}$, hats ($\widehat{\cdot}$) indicate complex Fourier coefficients, $\vec{k}^r$ is a wave-vector in the resolved scales, and subscript $\square$ implies that a “quadrangle” wave-vector $\vec{l}$ is involved in each term such that $\vec{k}^r$, $\vec{p}_{\square}$, $\vec{q}_{\square}$, and $\vec{l}$ form a quadrangle $\vec{l} = \vec{k}^r - \vec{p}_{\square} - \vec{q}_{\square}$, illustrated in Fig. 1.b. Equation 3 shows that the content of a single RS Fourier mode $\hat{\omega}_i(\vec{k}^r)$ due to nonlinear second-order chemical reactions is given by the double sum of all “quadratic” interactions involving Fourier modes $\widehat{K}_j(\vec{p}_{\square})$, $\widehat{\theta}_m(\vec{q}_{\square})$, and $\widehat{\theta}_n(\vec{l})$, defined by $\widehat{\square}(\vec{k}^r, \vec{p}_{\square}, \vec{q}_{\square}; \vec{l})$. Within the double summations in the RHS of Eq. 3 the fully-resolved quadratic interactions are identified as $\widehat{\square}(\vec{k}^r, \vec{p}_{\square}^r, \vec{q}_{\square}^r; \vec{l}^r)$, while all other RS-SFS interactions are given by quadrads with at least one SFS wave-vector $\vec{p}_{\square}^s$, $\vec{q}_{\square}^s$, and/or $\vec{l}^s$.

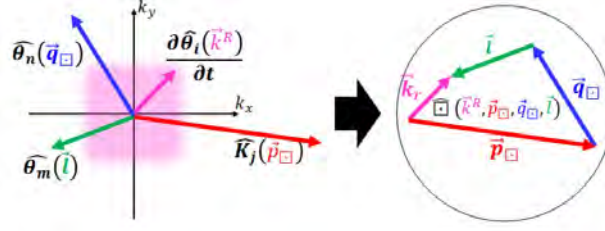


Figure 1: Fundamental quadratic form of chemical nonlinearities associated with second-order reactions in Eq. 3. The nonlinear contribution to the chemical source term $\widehat{\omega}_i(\vec{k}^r)$ is given by the summation of all quadrads representing the fundamental interaction between wave-vectors $\vec{k}^r$, $\vec{p}_\square$, $\vec{q}_\square$, and $\vec{l} = \vec{k}^r - \vec{p}_\square - \vec{q}_\square$ in multidimensional Fourier space that defines a quadrad $\widehat{\square}(\vec{k}^r, \vec{p}_\square, \vec{q}_\square; \vec{l})$.

The mathematical framework described here for third-order nonlinearities provides a novel description to identify and quantify inter-scale interactions between key spectral subregions. Whereas in [7] we addressed the advective nonlinearities, here we address the chemical nonlinearities associated with second-order reactions in the filtered species transport equations. The framework is generalizable to other types of nonlinearities and even higher-order nonlinearities within the governing equations of reacting flows. Using the framework developed in this section, we identify the dynamically dominant quadratic interactions in Eq. 3 for the evolution of resolved-scale species concentration per unit volume and isolate the SFS content within the dynamically dominant couplings in Fourier and physical space.

3. Reduced Physics Simulations: Flame-Vortex Interactions

The numerical simulation is performed using a 2D rectangular domain illustrated in Fig. 2 with length $L_y = 4D$ and $L_x = 16D$ in the cross-stream and streamwise directions, respectively, where D is the vortex diameter. An inflow of unburnt n -heptane/air mixture with $\phi = 0.95$, $T_u = 300$ K, and $P = 1$ atm is injected in the streamwise direction, the burnt products exit through the outflow plane, and periodic boundary condition is enforced in the cross-stream direction. The thermochemical conditions in the unburnt mixture are selected according to those used in the experiments of moderate Ka turbulent premixed flames [9].

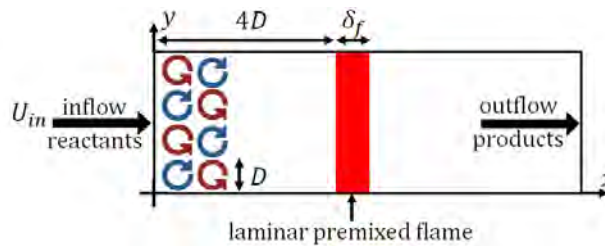


Figure 2: Schematic of computational domain for simulation of 2D flame-vortex interactions.

A planar n -heptane/air laminar premixed flame with the same unburnt conditions is initially placed 4 vortex diameters away from the inflow boundary. The planar laminar flame thickness and flame burning speed are computed to be 0.5 mm and 39 cm/s, respectively, using the chemical

kinetic models described in Sec. 3.1. The laminar flame thickness is defined as the distance between the iso-levels of 5% and 85% of progress variable, $C = (T - T_u)/(T_b - T_u)$, where T_u and T_b are the temperatures in the unburnt and burnt mixture, respectively.

An array of counter-rotating vortex pairs is continuously injected in the inflow boundary using time varying Dirichlet boundary conditions for the velocity components. The array of vortices is kinematically constructed from a combination of two sine waves aligned with the cross-stream and streamwise directions resulting in an array of single-scale vortices with zero averaged velocity. More details regarding the construction of the array of vortices is found in [6]. A streamwise injection velocity $U_{in} = 54$ cm/s is added to the kinematic velocity field, such that U_{in} is set to match the statistically averaged flame burning speed from the simulation. The same vortices that are injected in the inflow boundary are placed in the reactants side of the computational domain ($x \leq 4D$) as initial condition for the simulation.

The vortex length scale, l_v is computed as the square root of the surface area within the iso-level of 30% of the maximum vorticity divided by $\pi/4$, which captures more than 90% of the total vortex circulation. As such, l_v represents the effective diameter of the vortex core where most of the circulation is contained (63% of the vortex diameter in the sinusoidal velocity distribution described above, $l_v = 0.63D$). Accordingly, the vortex velocity scale, u_v , characterizes the strength of the vortex core, and is computed as the circulation contained within the same vorticity iso-level divided by its arclength.

The simulation is carried out employing vortices representative of inertial-range eddies in the reactant side of turbulent premixed flames in the thin reaction zone regime (Karlovitz number of order one). The vortex length scale is significantly larger than the flame length scale ($l_v/\delta_f = 10$) to elucidate the distinct characteristics of vortices and the premixed flame front in scale (Fourier) space. Therefore, the simulation is design such that $l_v/\delta_f = 10$, $u_v/S_L = 0.5$, $Ka = 2.5$, and $Re_v = 250$, where the reported values are computed after the vortices travel from the inflow plane to the vicinity of the flame front on the unburnt side. This simulation is labeled as “weak” flame-vortex interaction in our previous works [6, 7].

To use Fourier space quantitatively in non-periodic directions, we applied a recently developed “signal discontinuity pollution removal procedure” [10] to all variables of interest with $\alpha = 0.85$ and considering only C^0 boundary discontinuities. As a result, the maximum signal modification parameter and residual boundary discontinuity are smaller than 5% and 30%, respectively. The reader is referred to the original manuscript [10] for more details regarding the procedure and the outcome of the procedure is described in [7].

3.1 Computational Methodology

The 2D flame-vortex interaction simulations are performed using the NGA code [11], where unsteady Navier-Stokes equations under the low Mach number approximation and the transport equations for temperature and mass fractions of the various species are solved simultaneously. The scalar equations are discretized using the BQUICK scheme, which ensures that the physical bounds of appropriate quantities are numerically preserved throughout the simulation without adding significant artificial diffusion [12]. A recently-developed computationally-efficient, semi-implicit, iterative method is used for the time-integration of chemical source terms of gas-phase species [13]. This method has been shown to be free of lagging errors, as efficient as an explicit time-integration per time step, and capable of using large, stable time step sizes [14].

A semi-detailed chemical kinetic model containing 35 species and 217 reactions (forward and backward reaction counted separately) [15] is used. The reader is referred to [15, 16] for complete details about the chemistry reduction procedure and chemical model validation. This chemical model has been used in moderate Ka turbulent premixed flame simulations to reduce the computational cost [15].

The computational mesh is non-uniform in streamwise direction and uniform in cross-stream direction, being most refined in the vortex and flame regions in streamwise direction and stretched towards the outflow boundary. At least 80 grid points are used to resolve the vortex length scale in the reactants side and 20 grid points within the flame front thickness. The numerical simulation is shown to give the same results when twice as many grid points are used in both directions and the time step size is $1 \mu s$ to accurately resolve the chemical time scales.

4. Fourier-Physical Space Structure of Chemical Species

The range of resolved-scales determined as per [6] is used to separate RS and SFS content in both physical and Fourier space for all chemical species within the semi-detailed chemical kinetic model of this work. The relative variance (*i.e.*, energy) content in RS and SFS for each chemical species is quantified by computing the “filtered variance content” as the sum of squared Fourier coefficients within and outside, respectively, of the RS wavenumber cutoffs ($k_{RS,x}$ and $k_{RS,y}$). This quantitative classification indicates that 25 out of the 35 chemical species are mostly *not* resolvable by the LES grid (less than 20% RS variance content), while only 4 and 6 chemical species are partially subfilter and mostly resolvable, respectively. The mostly resolvable species are characterized by combustion products (*e.g.*, ρY_{H_2O} and ρY_{CO_2}) and reactants (*e.g.*, ρY_{O_2} and $\rho Y_{C_7H_{16}}$), while partially subfilter and subfilter species represent combustion intermediates, such as ρY_{HCO} .

We aim to identify and contrast the structural features of the three classes of chemical species described above. Figures 3.a-c show normalized ρY_i at $y/D = 2$ for resolved, partially subfilter, and subfilter chemical species, respectively. Figures 3.d-f present 2D instantaneous iso-contours of a characteristic resolved ($\rho Y_{C_7H_{16}}$), partially subfilter (ρY_{CO}), and subfilter (ρY_{CH_2O}) chemical species, respectively. The bottom and top halves of Fig. 3.d-f show the original and RS filtered signals, where the two dotted black curves indicate the RS flame front defined by RS progress variable iso-levels of 5% and 85%. Figures 3.g-i show 2D iso-contours of the logarithmic squared Fourier coefficients of the three characteristic chemical species, where the wavenumber axes are normalized using the planar laminar flame thickness so that the solid red and magenta lines indicate the flame-scale box ($k_{x,y}\delta_f/\pi = \pm 1$) and the RS cutoff scales defined in [6].

Chemical species mostly resolvable by the LES grid show an overall “jump” in ρY_i from reactants to products over the corrugated flame front associated with creation of products or consumption of reactants (Figs. 3.a and d). In contrast, chemical species that have most of their variance content at subfilter scales show an overall “peak” in ρY_i associated with creation and consumption of combustion intermediates within the corrugated flame front (Figs. 3c and f). The chemical species partially subfilter have a combination of the overall “jump” and “peak” structures in ρY_i (Figs. 3b and e), which represents combustion intermediates that undergo creation within the flame front and consumption over a large scale oxidation layer in the burnt products side of the mixture. The top halves of Fig.3d-f indicate that the large scale structural features of chemical species are readily identified in the RS filtered signals. These include the RS corrugated flame front, which is less corrugated and thicker, with corresponding lower flame-normal gradients within the front,

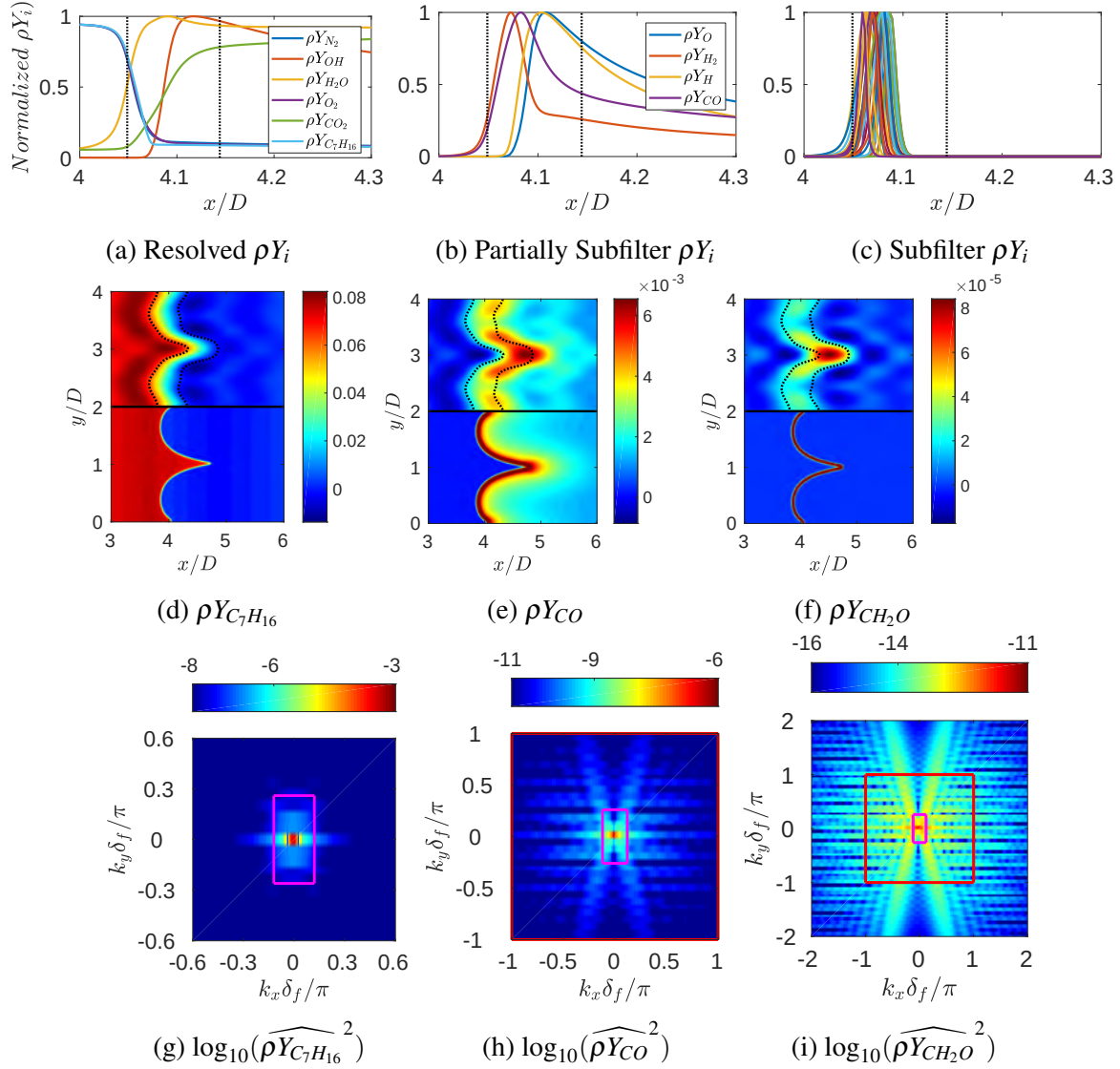


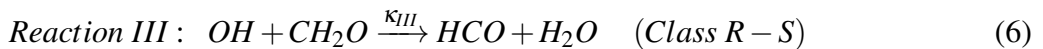
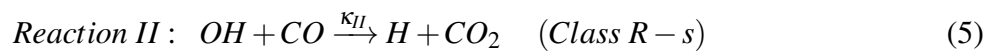
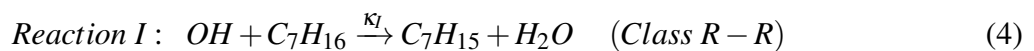
Figure 3: (a-c) Normalized ρY_i at $y/D = 2$ for resolved, partially subfilter, and subfilter chemical species, respectively. (d-f) Physical space iso-contours of a characteristic resolved ($\rho Y_{C_7H_{16}}$), partially subfilter (ρY_{CO}), and subfilter (ρY_{CH_2O}) chemical species, respectively (all in kg/m^3). Bottom and top halves show the original and RS filtered signals, respectively. (g-i) Logarithmic squared Fourier coefficients of the three characteristic chemical species.

than the full field which contains the small-scale features with the SFS motions. In particular, the RS flame cusps capture only the large-scale structural content of ρY_i . For ρY_{CH_2O} , the RS signal has significantly smaller magnitude compared with the original signal, as ρY_{CH_2O} has its variance mostly subfilter.

In Fourier space, resolved species show magnitude of Fourier coefficients with rapid reduction in energy from larger towards small scales, so that most of its variance content is at large scales resolved by the LES grid (magenta box in Fig. 3g). Subfilter chemical species display a much slower decay of Fourier content along the star-like pattern that extends up to $k_{x,y}\delta_f/\pi \approx \pm 2$, which is associated with the reaction zone thickness of the premixed flame where the concentration of most combustion intermediate species peaks. Partially subfilter chemical species display a slower rate of reduction from large to small scales extending up to $k_{x,y}\delta_f/\pi \approx \pm 1$.

5. Classification and Identification of Key Chemical Reactions

The understanding of the kinematic relationships between physical and Fourier space of the chemical species is now extended to classify distinct inter-scale couplings through the finite-rate chemistry source terms in the RHS of each species species transport equations. We classify the 217 chemical reactions based on the RS variance content of the participating species, which were classified in Sec. 4 as mostly resolvable, partially subfilter, and subfilter. The temperature-dependent rate constant of reaction is not included in the classification since all κ_j were observed to be mostly resolved (*i.e.*, $\geq 85\%$ RS variance), as temperature is mostly resolved by the LES grid [6]. To simplify the notation henceforth, we indicate mostly resolvable, partially subfilter, and subfilter variance content of kinematic variables by letters R, s, and S, respectively. As per Sec. 2, we focus our analysis in second-order reactions that represent 88% of the number of reactions within the chemical model of this work and comprise the most common type of chemical nonlinearities in premixed combustion [8]. From these second-order chemical reactions, three physically-relevant reactions with distinct reaction classifications are selected to investigate and contrast its dynamically dominant inter-scale couplings,



where reactions are numbered from *I* to *III*, as indicated in the subscripts of each reaction rate functions, $\kappa_j(T)$. The classification of each reaction is indicated within parenthesis. Reaction *I* (Eq. 4) is part of the initiation step for the oxidation of *n*-heptane (C_7H_{16}) by hydroxyl radical (OH), which begins the hydrocarbon oxidation process. Reaction *II* (Eq. 5) is a chain propagating step associated with conversion of CO to CO_2 and is the highly exothermic portion of any hydrocarbon oxidation system [8]. Reaction *III* (Eq. 6) is an important step that all carbon atoms in alkyl hydrocarbons go through before conversion to CO through the oxidation of formaldehyde (CH_2O) to formyl radical (HCO), which occurs within the thin reaction zone of a premixed flame. The finite-rate chemistry source terms of these three chemical reactions involve two resolved variables (κ_j and ρY_{OH}) and the third variable can be resolved ($\rho Y_{C_7H_{16}}$), partially subfilter (ρY_{CO}), or

subfilter (ρY_{CH_2O}), so that reactions *I* to *III* are classified as R-R, R-s, and R-S, respectively. We anticipate that the knowledge gained from analyses of these three characteristic chemical reactions may be extended to other reactions within these classes of reactions in the chemical mechanism.

6. Dynamically Dominant Chemical Inter-Scale Couplings and Subfilter-Scale Content

In this section we identify dynamically dominant inter-scale couplings by grouping quadrads according to specific features and we extract the corresponding SFS content of K_j , θ_m , and θ_n associated with dynamically dominant RS-SFS couplings. The reader is referred to [7] for a detailed description of the method to identify dynamically dominant nonlinear interactions and only a brief description is provided below.

The dynamical evolution of a single RS Fourier mode due to a chemical source term $\hat{\omega}_i(\vec{k}^r)$ occurs within a double summation over all Fourier modes $\vec{p}_\square$ and $\vec{q}_\square$ in the RHS of Eq. 3, which results in a list of quadrads representing all the chemical inter-scale interactions among Fourier modes $\vec{k}^r$, $\vec{p}_\square$, $\vec{q}_\square$, and $\vec{l} = \vec{k}^r - \vec{p}_\square - \vec{q}_\square$. The sum of all quadrads associated with a single RS Fourier mode $\vec{k}^r$ recovers the chemical nonlinearity in the LHS of Eq. 3. The list of quadrads associated with a single RS Fourier mode $\vec{k}^r$ are ranked and sorted using a “correlation criterion” as defined in [7] which provides information regarding the magnitude and phase correlations, respectively, between each quadrad and the sum of all quadrads. The first quadrads in the sorted list are more dynamically important relative to other quadrads. To down select the dynamically dominant quadrads within the sorted list, a “truncation criterion” as defined in [7] is defined such that the partial sum of the truncated set of quadrads recovers a specified (large) percentage of the total sum. The choice of the truncation criterion provides a reduction in the number of dynamically dominant SFS wave-vectors $\vec{p}_\square^{ds}$, $\vec{q}_\square^{ds}$, and $\vec{l}^{ds}$ for a given $\vec{k}^r$ mode, but also a reduction in the energy content in the chemical nonlinearity with the reduced subset of dynamically dominant SFS modes. To achieve a significant reduction in the degrees of freedom in the chemical nonlinearities, we select truncation criteria of 0.2, 0.5, and 0.5 for reactions *I*, *II*, and *III*, respectively.

6.1 Dynamically Dominant Sub-Filter Scale Content: Fourier Space

The dynamically dominant SFS content in Fourier space of each participating variable within the chemical nonlinearities $\widehat{K}_j(\vec{p}_\square^{ds})$, $\widehat{\theta}_m(\vec{q}_\square^{ds})$, and $\widehat{\theta}_n(\vec{l}^{ds})$ is shown in Fig. 4 for chemical reactions *I-III* (Eq. 4-6). Consistent with Fig. 3, we plot 2D iso-contours of the logarithmic squared Fourier coefficients of each variable, but only the dynamically dominant SFS modes are colored in the bottom half of each figure (making use of the symmetries discussed in Sec. 4). In the bottom halves, the resolved scales and nondominant SFS are indicated by white.

The dynamically dominant SFS modes selected by the method are shown to represent a negligible fraction of SFS modes for the R-R chemical reaction *I* in Figs. 4a-c. Since K_I , ρY_{OH} , and $\rho Y_{C_7H_{16}}$ are mostly resolved variables in reaction *I* (Eq. 4), the RS-RS nonlinear contribution dominates the nonlinear term. Therefore, the chemical source term associated with reaction *I* is mostly resolved by the LES grid and RS-SFS couplings have significantly small contribution. As such, we will not further analyze the dynamically dominant SFS content of chemical reaction *I*.

The same is not true for R-s and R-S chemical reactions *II* and *III*, respectively, shown in Figs. 4d-i. For these chemical reactions, the dynamically dominant SFS spectral modes extend from the SFS boarder with the RS cutoff wavenumbers into the higher wavenumbers (small scales)

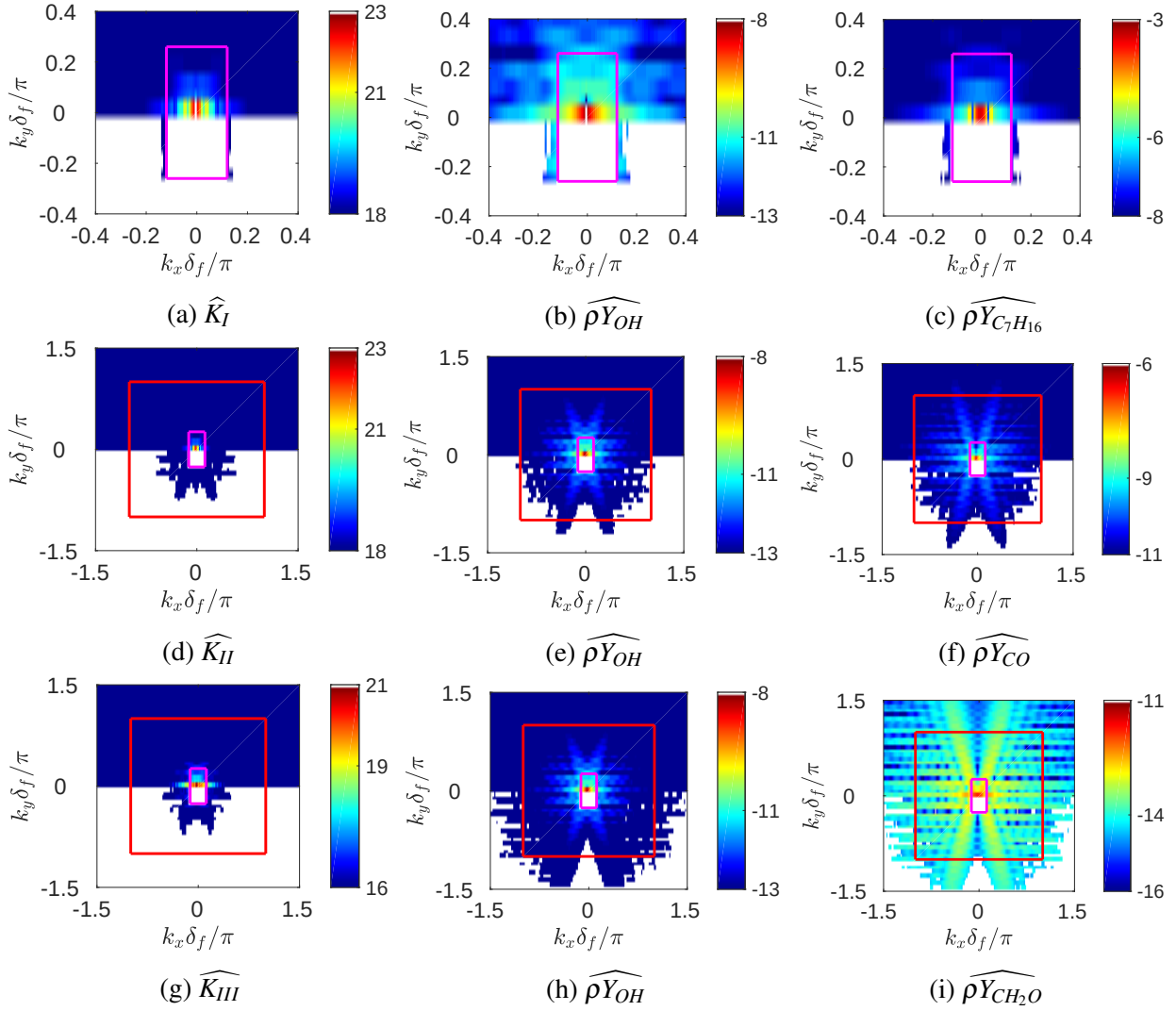


Figure 4: Iso-contours of the logarithmic squared Fourier coefficients of variables within reactions (a-c) *I*, (d-f) *II*, and (g-i) *III* (Eq. 4-6) classified as R-R, R-s, and R-S, respectively. Bottom halves of each figure show colored dynamically dominant SFS modes and white iso-level indicate resolved or nondominant subfilter scales. (a) $\log_{10}(\widehat{K}_I^2)$; (b) $\log_{10}(\widehat{\rho Y_{OH}}^2)$; (c) $\log_{10}(\widehat{\rho Y_{C_7H_{16}}})^2$; (d) $\log_{10}(\widehat{K}_{II}^2)$; (e) $\log_{10}(\widehat{\rho Y_{OH}}^2)$; (f) $\log_{10}(\widehat{\rho Y_{CO}}^2)$; (g) $\log_{10}(\widehat{K}_{III}^2)$; (h) $\log_{10}(\widehat{\rho Y_{OH}}^2)$; (i) $\log_{10}(\widehat{\rho Y_{CH_2O}}^2)$.

following the star-like pattern described in detail in [6]. For chemical species, the dynamically dominant SFS modes extend much farther from the RS cutoff wavenumbers, up to $|\vec{k}|\delta_f/\pi = 1.1$ and 1.5 for R-s reaction *II* (Fig. 4e-f) and R-S reaction *III* (Fig. 4h-i), respectively. The dynamically dominant SFS modes of the modified reaction rate functions $K_{II}(T)$ (Fig. 4d) and $K_{III}(T)$ (Fig. 4g) do not extend as far in Fourier space compared to the associated chemical species and are contained within $|\vec{k}|\delta_f/\pi \leq 0.8$

For chemical species *not* well resolved by the LES grid, such as ρY_{CO} and ρY_{CH_2O} , there is a clear correlation between the location of the dynamically dominant SFS modes (bottom half of each figure) and the more energetic Fourier modes within the star-like pattern (upper half of each figure). This correlation is less clear for variables mostly resolved, such as K_{II} , K_{III} , and ρY_{OH} ; here the more energetic Fourier modes are within the RS cutoff wavenumbers and the dynamically dominant SFS modes extend farther into the higher SFS wavenumbers. The locations of the dynamically dominant SFS modes of ρY_{OH} are shown to follow the SFS locations of participating species ρY_{CO} in Fig. 4e or ρY_{CH_2O} in Fig. 4h. The same is not true for K_{II} and K_{III} , where the dynamically dominant SFS modes follow the lower wavenumber content (large subfilter scales) of the star-like pattern. This indicates that there is greater correlation between the dynamically dominant small scales of species within R-s and R-S reactions *II* and *III*, respectively, but the associated reaction rate functions contribute through modulations at the largest scales within the SFS star-like pattern.

6.2 Dynamically Dominant Sub-Filter Scale Content: Physical Space

Figure 5a-b shows the dynamically dominant SFS content in physical space of $(K_{II})^{ds}$ and $(\rho Y_{CO})^{ds}$ for R-s reaction *II*; Fig. 5c-d shows $(K_{III})^{ds}$ and $(\rho Y_{CH_2O})^{ds}$ for R-S reaction *III*. The complete SFS signal of each variable is shown in the bottom halves of each figure for comparison. Figures 6a-b shows the dynamically dominant SFS of ρY_{OH} in reactions *II* and *III*, and its entire SFS content is shown in 6c. The two-dimensional mean value of the respective original signals is added for a direct comparison and the two dotted curves indicate the RS flame front defined by RS progress variable iso-levels of 5% and 85%. The “ghost ripples” observed in Figs. 5 and 6 are a result of the sharp filters employed to obtain the physical-space signals, referred as “ringing effect”, which may be minimized (not shown here) with the use of smooth Fourier space filters as representative of true smooth filters in operational LES.

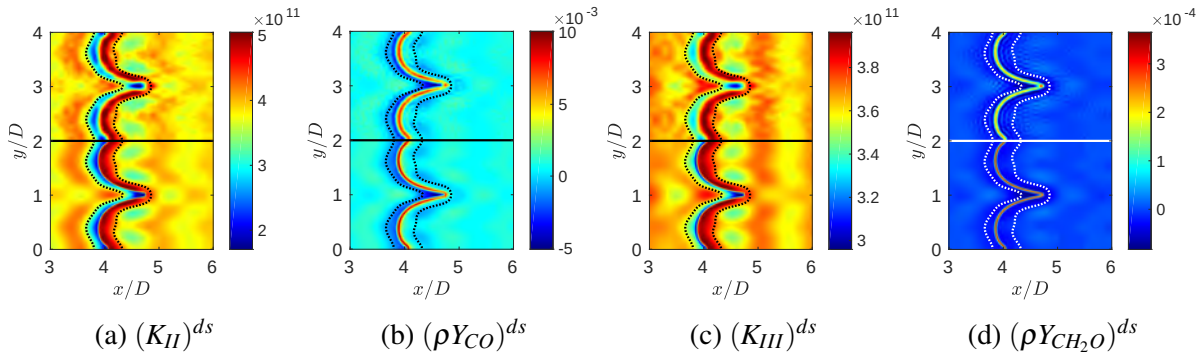


Figure 5: Iso-contours of species mass fraction (a) K_{II} and (b) ρY_{CO} filtered at dynamically dominant SFS for R-s reaction *II* (Eq. 5) and (c) K_{III} and (d) ρY_{CH_2O} filtered at dynamically dominant SFS for R-S reaction *III* (Eq. 6). Entire SFS content is shown in bottom halves of each panel.

The dynamically dominant SFS content in physical space of chemical nonlinearities is shown to be localized in the regions with largest gradients over the reaction zone within the flame front. For variables mostly resolved by the LES grid, such as K_{II} (Fig. 5a), K_{III} (Fig. 5c), and ρY_{OH} (Fig. 6), the SFS content within the corrugated flame fronts has a double-banded structure in physical space surrounding the reaction zone, similar to the advective nonlinearity for $(\rho h)^{ds}$ in [6]. As the dynamically dominant SFS modes extend farther from the RS cutoff wavenumbers in the R-S reaction compared to R-s reaction, the double-banded structure recovers most of the smallest-scale gradients within the reaction zone observed in the entire SFS for $(K_{III})^{ds}$ and $(\rho Y_{OH})_{III}^{ds}$. In contrast, $(\rho Y_{CH_2O})^{ds}$ in Fig. 5d is a variable mostly subfilter and shows a much thinner high-gradient structure that peaks in the reaction zone within the RS flame front, which is similar to the advective dominant SFS content of $(\rho Y_{HCO})^{ds}$ in [6]. The dynamically dominant SFS content of the partially subfilter variable $(\rho Y_{CO})^{ds}$ in Fig. 5b is shown to capture the overall double-banded structure surrounding the reaction zone but with smaller-scale content associated with its thickness and localization around the cusps compared to the variables mostly resolved described above. In general, the truncation of dynamically dominant SFS modes removes small-scale features in the SFS signals, without fundamental modification in the structure of the SFS signal.

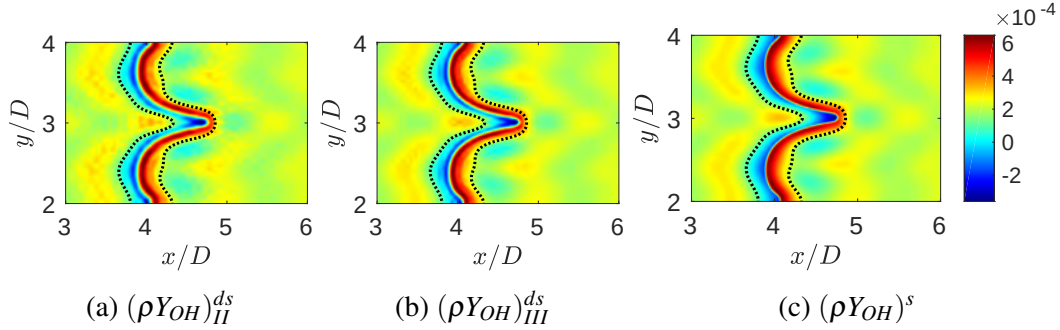


Figure 6: Iso-contours of species mass fraction ρY_{OH} filtered at dynamically dominant SFS for reactions (a) II and (b) III indicated by subscripts II and III , respectively. Entire SFS content of ρY_{OH} is shown in (c).

7. Conclusions

In this work, we make use of the previous analyses presented in [6, 7, 10] that relate spectral content to physical space content of key variables in premixed turbulent combustion to characterize nonlinear chemical inter-scale couplings between RS and SFS motions. Chemical species mostly resolvable by the effective LES grid show an overall “jump” from reactants to products over the corrugated flame front in physical space and magnitude of Fourier coefficients with fast decay of energy towards small scales. In contrast, chemical species that have most of its variance content at subfilter scales show an overall “peak” within the reaction zone in physical space and display a much slower decay of Fourier content along the star-like pattern towards high wavenumbers. The chemical species partially subfilter are shown to have a combination of the overall structural characteristics of resolved and subfilter species described above.

The chemical inter-scale interactions between distinct chemical species are identified and classified based on the RS variance content of the participating species (Sec. 5). We show that most of

the chemical reactions within the semi-detailed chemical kinetic model employed in this work (Sec. 3) involve interactions with chemical species *not* resolvable by the LES grid, so that the dynamical evolution of these chemical species is dominantly subfilter-scale and therefore contrary to the classical LES framework and associated SFS modeling strategies. We analyzed three physically-relevant chemical reactions (Eq. 4-6) and contrasted the dynamically dominant SFS content using the methodology developed in [7] and extended here for chemical nonlinearities (Sec. 6).

The dynamically dominant SFS modes for chemical reactions with resolvable participating species (R-R chemical reaction) comprise a negligible fraction of SFS modes, as the RS-RS nonlinear contribution dominates the entire nonlinear term. Therefore, the evolution of the resolved scales due to the R-R chemical reaction is mostly resolved by the LES grid and RS-SFS couplings have significantly small relative energy content. In contrast, chemical reactions involving species not resolvable by the LES grid (R-s and R-S reactions) show a larger energy content within RS-SFS couplings.

The extent to which the dynamically dominant SFS modes of chemical reactions *II* and *III* extend much farther away in spectral space from the RS cutoff wavenumbers reflects the dominant SFS dynamics associated with the RS evolution of chemical species within the reaction zone. As per discussion of Sec. 1, key dynamics underlying the function of combustion devices are almost always dominantly in the unresolvable SFS motions, as now evident from the dynamically dominant SFS content of chemical nonlinearities.

The physical-space counterpart of the dynamically dominant SFS Fourier content associated with R-s and R-S chemical reaction is shown (Sec. 6.2) to capture the overall structure of the SFS signals without the smallest-scale gradients within the flame cusps and the reaction zone. As the dynamically dominant SFS modes extend farther from the RS cutoff wavenumbers in the R-S reaction compared to R-s reaction, the physical-space counterpart recovers most of the smallest-scale gradients within the reaction zone. The extent the small-scale features in the dynamically dominant SFS signals are removed is directly related to the classification of each nonlinearity within its inter-scale coupling. These results are potentially valuable in the design of new SFS modeling strategies for key variables in LES of premixed turbulent combustion wherein only RS data on the LES grid are available, in principle.

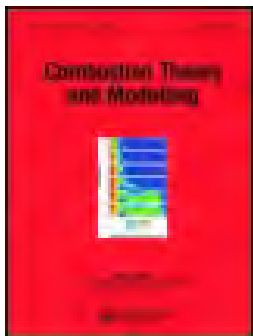
8. Acknowledgements

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Assessment of disparities in estimating filtered chemical reaction rates in LES using DNS of turbulent premixed flames

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Assessment of disparities in estimating filtered chemical reaction rates in LES using DNS of turbulent premixed flames

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An *a priori* assessment of the disparities in estimating the filtered chemical reaction rates in Large-Eddy Simulations (LES) is performed using two Direct Numerical Simulations (DNS) of turbulent premixed flames. DNS of turbulent premixed flames with the same turbulent Reynolds number but different Karlovitz numbers are carried out using semi-detailed finite-rate chemistry to represent different flame conditions. The sensitivities of the differences between the LES-filtered chemical reaction rates and those constructed using LES-filtered thermochemical variables (referred to as ‘disparity’ below) to reaction types, species characteristics, and flame conditions are systematically investigated. The sensitivity to reaction type is examined by quantifying the disparity in the filtered temperature-dependent reaction rate constant as a function of kinetic rate parameters. The sensitivity to species characteristics is examined by quantifying the disparity in the filtered law of mass action as a function of species chemical time scales and formation location. The sensitivity to flame conditions is examined by contrasting the results obtained from the two simulations. These disparities are compared to characterise distinct reaction/species categories with different levels of disparity in estimating the filtered chemical reaction rates in LES. For both flames, it is found that the rate constants estimated using LES-filtered temperature severely under-predict the LES-filtered rate constants, especially for reactions with high activation energies. The law of mass action estimated using LES-filtered species concentrations severely over-predicts the LES-filtered law of mass action, especially for reactions involving reactants with small chemical time scales. The combined effects are found to lead to considerable differences between the LES-filtered chemical reaction rates and those constructed using LES-filtered thermochemical quantities for all reaction types involving different classes of reactants typically included in combustion reaction mechanisms. The details and explanations of these differences provide test cases useful to the improvement of key LES modelling approximations.

Keywords: direct numerical simulation; large-eddy simulation; turbulent premixed flames; chemical reaction rates

1. Introduction

Large-Eddy Simulation (LES) has become a promising approach for engineering design and optimisation [1,2]. In LES, larger-scale, geometry-dependent flow features are resolved by the computational grid, and closure models are needed to describe the effects of smaller scales on large-scale flow evolution [1,3–5]. Specifically for LES of turbulent

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combustion, such models are needed for sub-filter Reynolds stresses, scalar fluxes and scalar variances, as well as chemical reaction (production/consumption) rates [2,3].

Over the past decades, research has focussed on the LES modelling of chemical reaction rates through a variety of turbulent combustion models including low-dimensional manifold-based models [6–8], thickened flame models [9], transported probability density function models [10,11], approximate de-convolution methods [12,13], conditional moment closure models [14], and linear eddy models [15,16]. These models are generally needed because variations in temperature (T) and species concentrations (C) are typically not adequately resolved so that the chemical reaction rates computed directly using LES-filtered variables can differ significantly from the LES-filtered chemical reaction rates, $\bar{w}(T, C)$ [1], where bar is used to represent filtered quantities. These large differences are also due to the high degree of nonlinearity in the chemical reaction rate formulation, where for an irreversible reaction i , $\sum_{j=1}^{N_i} \nu'_j A_j \rightarrow \sum_{j=1}^{N_i} \nu''_j A_j$, the reaction rate is given by the ‘law of mass action’,

$$w_i(T, C) = k_i(T) \prod_{j=1}^{N_i} C_j^{\nu'_j}. \quad (1)$$

In the above equation C_j is the concentration of species j , ν'_j and ν''_j are the stoichiometric coefficients of species j on the reactants and products sides respectively and N_i is the total number of species involved in reaction i . $k_i(T)$ is the temperature-dependent rate constant expressed using the Arrhenius rate law in terms of the pre-exponential factor A_i , the temperature exponent n_i , and the activation energy $E_{a,i}$:

$$k_i(T) = A_i T^{n_i} \exp\left(\frac{-E_{a,i}}{R_u T}\right). \quad (2)$$

The disparity in the estimation of the filtered law of mass action (e.g. the cross correlation of reactant concentrations) has been the focus of early turbulent closure modelling for reacting flows (e.g. [17]). More recently in context with LES, disparities in estimating LES-filtered chemical reaction rates have been examined in previous studies using *a posteriori* analyses in LES [18] and *a priori* analyses based on DNS of turbulent flames [19]. In [18], LES was performed using the so-called ‘finite-rate chemistry approach’, suggesting a reasonable agreement with experimentally-measured ensemble-averaged velocity and temperature in turbulent premixed swirl-stabilised and bluff body-stabilised flames. On the other hand, the exact $\bar{w}_i(T, C)$ has been found to deviate substantially from the estimated values based on LES-filtered quantities in DNS of freely-propagating turbulent premixed flames, leading to different predictions of dominant chemical reaction pathways, especially at high Karlovitz numbers, Ka [19]. The extent of such deviations should, in principle, depend on the turbulent flame conditions (e.g. turbulent Reynolds number Re and Ka), reaction-specific rate parameters and chemical characteristics of the participating reactants in relationship to LES filter width.

Although the substantial deviations between the filtered reaction rates and those estimated from LES-filtered quantities have been acknowledged extensively in previous studies, for instance in the works stated above, how these deviations depend on the type of reactions, participating species and turbulent flame conditions remains unclear. This lack of clarity makes it difficult to develop improvements to reduce the error associated with the representation of the filtered highly nonlinear chemical reaction rates.

Our main objective is to quantify the sensitivities of the disparities between the true LES-filtered chemical reaction rates that LES attempts to model and those constructed using LES-filtered thermochemical quantities as a function of reaction type, species characteristics and flame conditions, using DNS-based *a priori* analysis. We perform DNS of two turbulent premixed flames with the same Re but different Ka to contrast the sensitivity to flame conditions. For each flame, we first investigate the sensitivity to reaction type by quantifying the disparity in filtered reaction rate constant as a function of kinetic rate parameters for all reactions in the chemical kinetic model used in the DNS. We then investigate the sensitivity to species characteristics by quantifying the disparity in the filtered law of mass action as a function of species chemical time scales and reactant formation location for all reactions. Finally, we combine these disparities to identify distinct reaction categories with different levels of disparity in estimating the filtered chemical reaction rates relative to those formed with LES variables. The implications of the identified categories of reactions on the level of disparities are expected to be generalisable to flames operating with other fuels under different conditions. The main focus of this work is not to confirm the large disparities in the estimation of reaction rates, but to systematically investigate the location, magnitude, origin and sensitivities of such disparities. We point out that the intent of this paper is not to develop new models to improve the prediction accuracy of LES with finite-rate chemistry, but rather to identify important disparities and sensitivities that can guide model improvements and be applied to assess new model developments.

2. Numerical set-up and methodologies

2.1. DNS setup

We performed DNS for two *n*-heptane/air turbulent premixed flames with finite-rate chemistry at the same Re but at Ka differing by over an order of magnitude and referred to as ‘low’ vs. ‘high’ Ka cases. The configuration used in both simulations is that of a statistically-planar flame, with inflow/outflow boundary conditions in the stream-wise direction and periodic boundary conditions in the span-wise directions, similar to [19].

A fully-premixed, slightly lean *n*-heptane/air mixture is injected at the inflow plane with unburnt temperature $T^u = 300$ K at 1 atm pressure and equivalence ratio $\phi = 0.95$ for both flames. Under these conditions the laminar flame thickness and burning velocity were computed to be $l_F = 3.15 \times 10^{-4}$ m and $s_L = 0.39$ m/s, respectively, using the chemical model described in Section 2.2. Each flame was generated by injecting isotropic turbulence at the inflow plane that was generated from a separate incompressible 3D turbulence simulation at stationary state with spectral forcing [20] with correlation integral scale L_0 quantified in Table 1 relative to l_F . No forcing is imposed in the turbulent flame simulations and turbulence was allowed to decay freely downstream as it interacts with the flame, thus avoiding potential artifacts from forcing. For each flame, the integral and Kolmogorov length scales and the rms turbulence velocity were computed at the intended mean flame location ($4L_0$ from the inflow plane) and are given in Table 1 by l_0 , η_0 , and u_0 , respectively. These quantities with subscript 0 were calculated from transverse direction velocity components in a separate DNS of non-reacting, incompressible decaying turbulence with the same inflow and computational domain as the actual flame simulation. The Reynolds number $Re_0 = u_0 l_0 / \nu_0$ and Karlovitz number $Ka_0 = (l_F \nu_0) / (s_L \eta_0^2)$ [21] computed at this location are provided in Table 1. Re_0 is chosen so that l_0 / l_F is at least 10 and 2 in the low and high Ka simulations, respectively. Under these conditions, both flames are expected to fall in the thin reaction zones regime.

Table 1. Simulation parameters for the two flames.

Parameter	Low-Ka flame	High-Ka flame
L_0/l_F	7.05	0.66
l_0/l_F	11.57	2.22
u_0/s_L	3.38	17.1
l_F/η_0	7.8	46.92
Re_0	329	321
Ka_0	7.23	261
Grid	$900 \times 500 \times 500$	$350 \times 500 \times 500$
Domain	$43L_0 \times 7L_0 \times 7L_0$	$40L_0 \times 15L_0 \times 15L_0$
Cost	1.34×10^5 CPU-hours	2.2×10^5 CPU-hours

The computational domain was designed to initially accommodate more than 5 inflow integral-scale (L_0) eddies in the transverse directions and $> 40L_0$ in the stream-wise direction. The computational grid was chosen to resolve well the flame and turbulence, with at least 10 grid points across the laminar flame thickness and with $k_{\max}\eta_0 \geq 1.5$ in each direction, where $k_{\max}$ is the maximum wavenumber resolved by the grid. To reduce the associated computational cost, the grid is stretched downstream of the flame on the products side. The domain sizes and grid resolutions used in both flame simulations are summarised in Table 1. To overcome the initial transient phase, both simulations were performed first using flamelet-based tabulated chemistry for more than 23 eddy-turnover times and later using finite-rate chemistry till the variations in joint probability density distributions of species concentration and temperature are diminished. The approximate computational cost associated with both flame simulations is also reported in Table 1.

2.2. Computational methodology

A semi-detailed chemical kinetic model for *n*-heptane/air laminar premixed flame containing 35 species and 217 reactions (forward and backward reactions counted separately) [22] was used, reduced from the original model with 149 species and 1651 reactions [23]. Complete details and chemical model validation are given in [22]. This chemical model has been previously used in detailed simulations of 2D flame-vortex interactions [24] and DNS of 3D high *Ka* turbulent premixed flames [22].

The turbulent flame simulations were performed using the NGA code [25], where the unsteady Navier-Stokes equations with the low Mach number approximation and the transport equations for temperature and species mass fractions are solved simultaneously. The scalar equations are discretised using the BQUICK scheme, which ensures that the physical bounds of appropriate quantities are numerically preserved without adding significant artificial diffusion [26]. A recently-developed computationally-efficient, semi-implicit, iterative method is used for the time-integration of chemical source terms [27]. This method has been shown to be free of lagging errors, is efficient as explicit time integration, and is capable of maintaining stability with relatively large time step sizes [28].

3. Results and discussions

3.1. *A priori* filtering of DNS results

The DNS results were filtered using a top-hat box spatial filter with a fixed filter width $\Delta = 2l_F = 630\mu\text{m}$ to represent the LES filter, similar to previous studies [12,19,29].

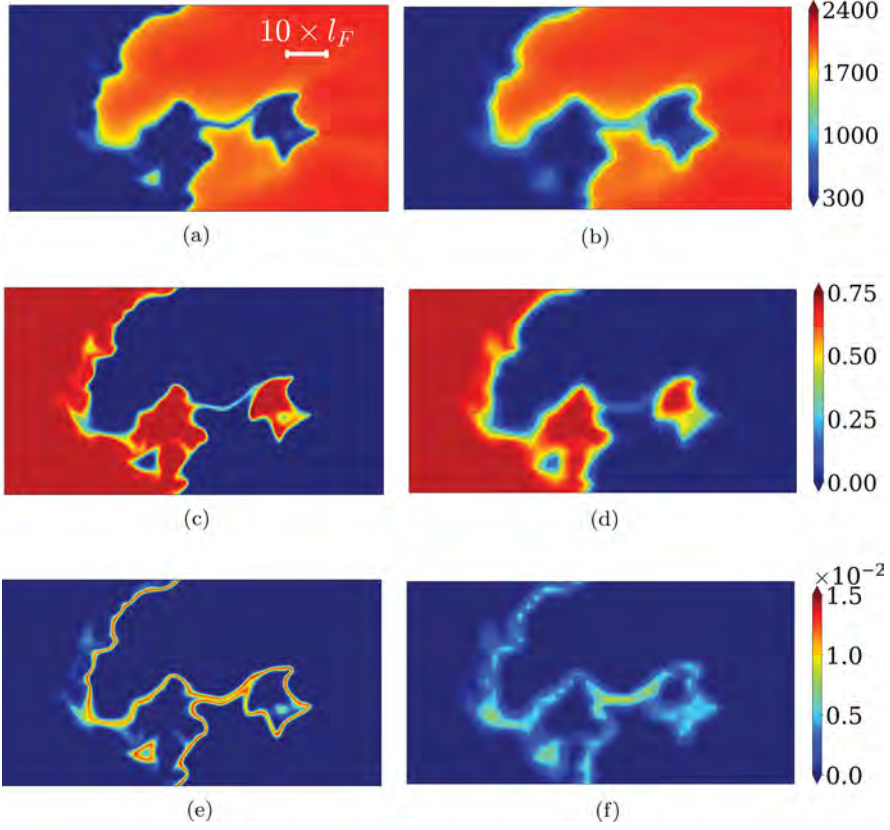


Figure 1. Original (left panel) and filtered (right panel) variables in the low- Ka flame. A length scale corresponding to $10 \times l_F$ is shown in panel (a). (a) T (K). (b) $\tilde{T}$ (K). (c) $C_{n-C_7H_{16}}$ (mol/m³). (d) $\bar{C}_{n-C_7H_{16}}$ (mol/m³). (e) C_{CH_2O} (mol/m³) and (f) $\bar{C}_{CH_2O}$ (mol/m³). (Color online)

Filtered DNS results are shown in Figure 1 for temperature and concentrations for fuel ($n-C_7H_{16}$) and an intermediate species (CH_2O) on a planar slice for the low- Ka case. Note that filtering is applied to species concentrations, $\bar{C}_j$, consistent with the Favre-filtered species mass fractions, $\tilde{Y}_j = \bar{\rho} \bar{Y}_j / \bar{\rho} = \bar{C}_j W_j / \bar{\rho}$, commonly used in turbulent combustion LES, where W_j is the molecular weight of species j . For the same reason, density-weighted Favre-filtering is used for temperature, $\tilde{T} = \bar{\rho} \bar{T} / \bar{\rho}$. Figure 1 shows that filtering thickens the flame structure and smears the high gradients to the filter scale; this is especially apparent with species formed within the flame such as CH_2O . Similar results are found in the high- Ka case, as shown in Figure 2.

3.2. Disparity in estimating the filtered reaction rate constants

Five representative reactions, listed in Table 2, were selected to quantify the disparities between the filtered reaction rate constants, $\bar{k}(T)$, and the estimated values using the Favre-filtered temperature, $k(\tilde{T})$. These reactions encompass a wide range of kinetic rate parameters (E_a and n) and represent a variety of categories of reactions in the entire chemical kinetic model that includes both exothermic and endothermic reactions, uni-molecular

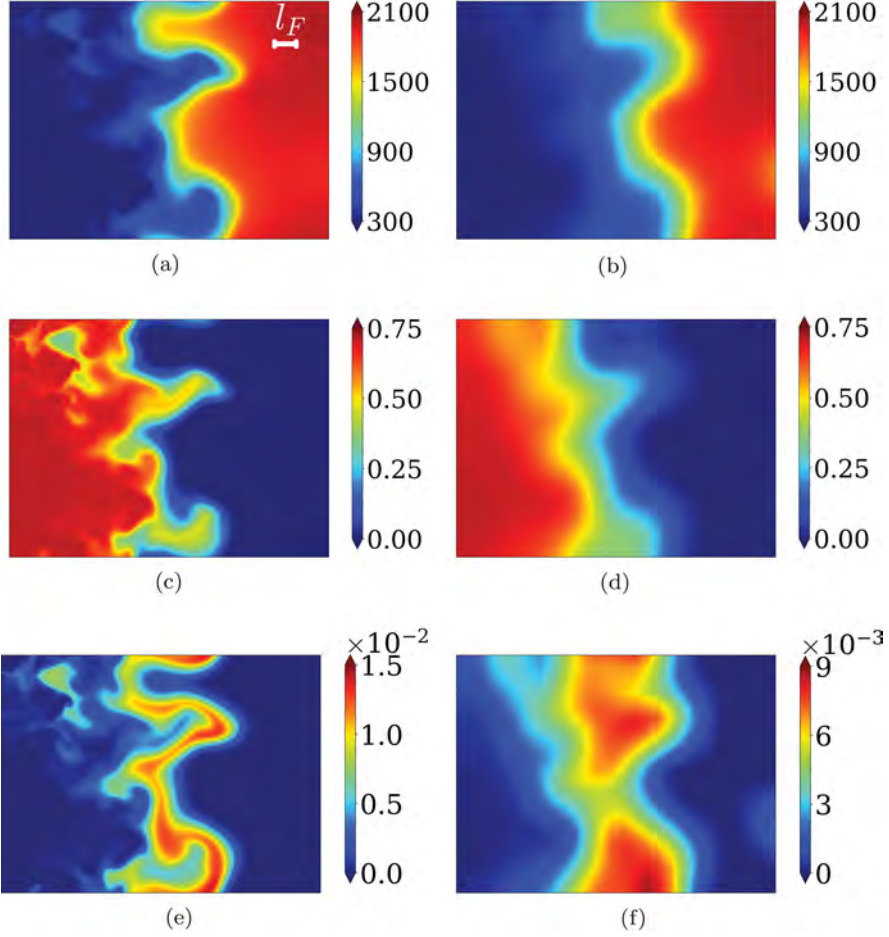


Figure 2. Original (left panel) and filtered (right panel) variables in the high- Ka flame. The laminar flame thickness l_F is indicated in panel (a). (a) T (K). (b) $\tilde{T}$ (K). (c) $C_{n-C_7H_{16}}$ (mol/m³). (d) $\bar{C}_{n-C_7H_{16}}$ (mol/m³). (e) C_{CH_2O} (mol/m³) and (f) $\bar{C}_{CH_2O}$ (mol/m³). (Color online)

fuel decomposition reactions, and bi-molecular oxidation reactions. Furthermore, the reactions include reactants from radicals to stable molecules that evolve with vastly different chemical time scales. Figure 3 compares the conditional mean profiles of $\bar{k}(T)$ and $k(\tilde{T})$ for these reactions over the Favre-filtered progress variable,

$$\tilde{\xi} = \frac{\tilde{T} - T^u}{T^b - T^u}, \quad (3)$$

where T^u and T^b are the constant temperatures in the unburnt reactants and burnt products. These profiles are normalised by the maximum value of $\langle \bar{k}(T) | \tilde{\xi} \rangle$.

For all reactions, the differences between $\langle \bar{k}(T) | \tilde{\xi} \rangle$ and $\langle k(\tilde{T}) | \tilde{\xi} \rangle$ are shown in Figure 3(a,b) to be within $0 \lesssim \tilde{\xi} \lesssim 0.8$, and tend to diminish towards high values of $\tilde{\xi}$ irrespective of the rate parameters. This is because the turbulent fluctuations are dampened due to increased viscous dissipation in the products (high $\tilde{\xi}$), so turbulent fluctuations are

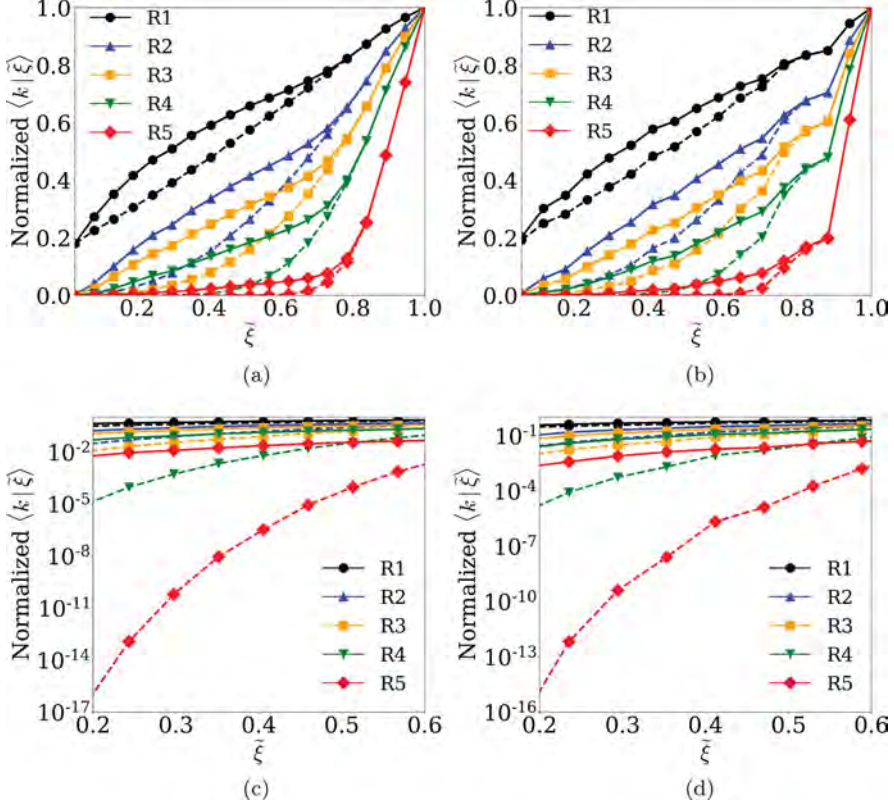


Figure 3. Conditional mean profiles of $\langle \bar{k}(T) | \tilde{\xi} \rangle$ (solid lines) and $\langle k(\tilde{T}) | \tilde{\xi} \rangle$ (dashed lines), normalised by the maximum value of $\langle \bar{k}(T) | \tilde{\xi} \rangle$, for reactions listed in Table 2. (a) Low- Ka case. (b) High- Ka case. (c) Low- Ka case; log-linear scale and (d) High- Ka case; log-linear scale. (Color online)

Table 2. Kinetic rate parameters for the representative reactions.

	n	$E_a \left[\frac{\text{kJ}}{\text{mol}} \right]$	Reaction
R1	1.2	-1.87	$\text{CH}_2\text{O} + \text{OH} \rightarrow \text{HCO} + \text{H}_2\text{O}$
R2	1.5	14.4	$\text{OH} + \text{H}_2 \rightarrow \text{H} + \text{H}_2\text{O}$
R3	2.8	9.46	$n\text{-C}_7\text{H}_{16} + \text{O} \rightarrow 2\text{-C}_7\text{H}_{15} + \text{OH}$
R4	-1.1	106	$\text{H} + \text{CO}_2 \rightarrow \text{OH} + \text{CO}$
R5	-17.6	504	$n\text{-C}_7\text{H}_{16} \rightarrow 1\text{-C}_5\text{H}_{11} + \text{C}_2\text{H}_5$

better resolved and filtering has less consequence. Despite the small differences between $\langle \bar{k}(T) | \tilde{\xi} \rangle$ and $\langle k(\tilde{T}) | \tilde{\xi} \rangle$ towards low $\tilde{\xi}$ where both terms approach zero, large relative differences could lead to significant errors in the estimation of filtered chemical reaction rates, because k is a multiplicative factor therein (Equation (1)). As shown in Figure 3(c,d) using log-linear scales, $\langle \bar{k}(T) | \tilde{\xi} \rangle$ and $\langle k(\tilde{T}) | \tilde{\xi} \rangle$ can differ by several orders of magnitude at low $\tilde{\xi}$, especially for reaction R5 with high E_a , where the participating species ($n\text{-C}_7\text{H}_{16}$) is predominantly in the unburnt gas (low $\tilde{\xi}$).

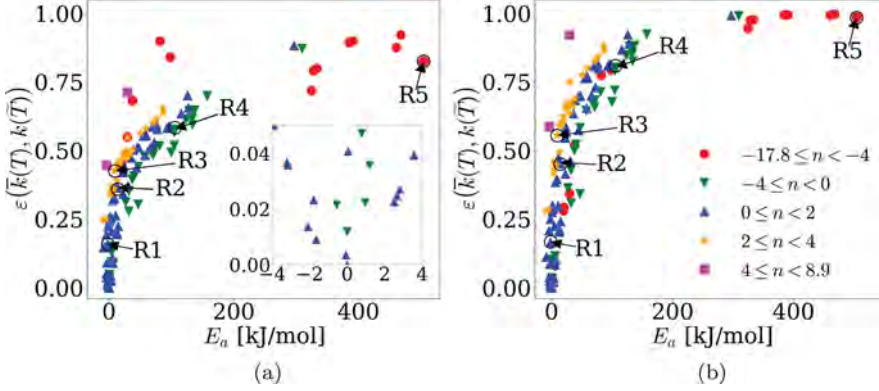


Figure 4. Difference metric ε evaluated between $\bar{k}(T)$ and $k(\tilde{T})$ for all reactions in both flames. The inset in the left panel shows an enlarged view of the difference metric for reactions with low E_a values. (a) Low- K_a case and (b) High- K_a case. (Color online)

For each test flame, $\bar{k}(T)$ is shown to be under-estimated by $k(\tilde{T})$ for all reactions. More specifically, reactions with higher E_a and n have larger deviations between $\langle \bar{k}(T) | \tilde{\xi} \rangle$ and $\langle k(\tilde{T}) | \tilde{\xi} \rangle$, for instance for reactions R4 and R5 where the dependence of k on T (Equation (2)) is exceptionally nonlinear with higher values of E_a and n . Comparing the two flames, the high- K_a case shows slightly larger deviations between $\langle \bar{k}(T) | \tilde{\xi} \rangle$ and $\langle k(\tilde{T}) | \tilde{\xi} \rangle$ for R1–R5. This is expected since the high- K_a flame has higher levels of turbulent temperature fluctuations at scales below the filter width.

To generalise these results to all reactions, we define the following L_2 norm-based global ‘difference metric’:

$$\varepsilon(f, g) = \sqrt{\frac{\int_V (f(\mathbf{x}) - g(\mathbf{x}))^2 d\mathbf{x}}{\int_V f(\mathbf{x})^2 d\mathbf{x}}}, \quad (4)$$

where the spatial integration is performed over a domain of interest V , selected to include the entire instantaneous turbulent flame defined as the region between instantaneous progress variables of 0.05 and 0.75 in our current analysis. Figure 4 shows $\varepsilon(\bar{k}(T), k(\tilde{T}))$ against E_a , computed for all reactions in the chemical kinetic model and grouped by different ranges of temperature exponent n for both flames.

Figure 4 shows that the difference metric ε has a strong dependence on E_a ; reactions with higher E_a exhibit larger ε . These high- E_a reactions are typically endothermic, for example the uni-molecular decomposition reaction R5 in Table 2, or reactions between stable molecules in a general chemical kinetic mechanism. On the other hand, many exothermic reactions such as R1, as well as general chain-propagating and chain-terminating reactions such as R2, have relatively small E_a values with correspondingly smaller values of ε (Figure 4), indicating that the filtered rate constants are better estimated using filtered temperature. This trend is observed consistently even for reactions with negative E_a (see inset in Figure 4), which are typically exothermic, can occur spontaneously, and exhibit decreasing rate constants with increasing temperature [30].

The difference metric ε exhibits weaker dependence on n and K_a than on E_a . n cannot be interpreted with reaction categories as they are typically determined to fit experimentally-measured rate constants with the Arrhenius rate law [30]. For reactions with similar E_a

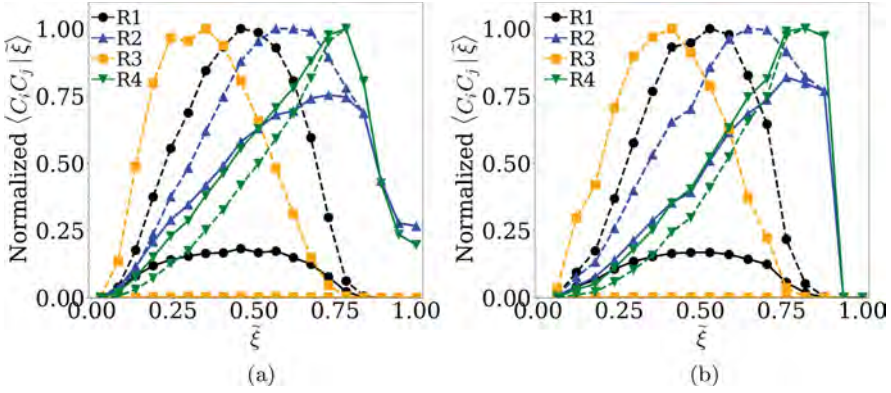


Figure 5. Conditional mean profiles of $\langle \overline{C_i} \overline{C_j} | \tilde{\xi} \rangle$ (solid lines) and $\langle \overline{C_i} \overline{C_j} | \tilde{\xi} \rangle$ (dashed lines), normalised by the maximum value of $\langle \overline{C_i} \overline{C_j} | \tilde{\xi} \rangle$, for R1–R4 in Table 2. (a) Low-Ka case and (b) High-Ka case. (Color online)

values, higher n magnitudes lead to slightly higher ε , as expected from Equation (2). Consistent with Figure 3, all reactions are found to have slightly higher ε in the high-Ka flame due to the larger temperature fluctuations.

3.3. Disparity in estimating the filtered law of mass action

The disparities between the filtered reactant concentration products $\overline{C_i} \overline{C_j}$ and the estimated value using filtered reactant concentrations $\overline{C_i} \overline{C_j}$ are quantified for the four bi-molecular reactions (R1–R4) from Table 2. Figure 5 shows a comparison between the conditional mean profiles of $\overline{C_i} \overline{C_j}$ and $\overline{C_i} \overline{C_j}$ over $\tilde{\xi}$ in both flames, normalised by the maximum value of $\langle \overline{C_i} \overline{C_j} | \tilde{\xi} \rangle$. The differences between $\langle \overline{C_i} \overline{C_j} | \tilde{\xi} \rangle$ and $\langle \overline{C_i} \overline{C_j} | \tilde{\xi} \rangle$ are within $0.1 \lesssim \tilde{\xi} \lesssim 0.8$, and tend to diminish towards high and low values of $\tilde{\xi}$, as expected. For both flames, reactions R1 and R3 exhibit particularly large deviations between $\langle \overline{C_i} \overline{C_j} | \tilde{\xi} \rangle$ and $\langle \overline{C_i} \overline{C_j} | \tilde{\xi} \rangle$, while reactions R2 and R4 exhibit much smaller deviations (within 40%). The high-Ka case shows only slightly larger deviations between $\langle \overline{C_i} \overline{C_j} | \tilde{\xi} \rangle$ and $\langle \overline{C_i} \overline{C_j} | \tilde{\xi} \rangle$ for R1–R4. This suggests that the degree of disparity in estimating the filtered law of mass action varies only marginally between the two flame conditions.

The disparity in estimating the filtered law of mass action varies significantly across different reactions (Figure 5) because the concentrations of different species are affected very differently by the filtering operation. More precisely, species with exceptionally high concentration gradients, typically associated with small chemical time scales, are much more affected than those with low concentration gradients, characterised by large chemical time scales. This is illustrated in Figure 6 for the reactants in reactions R3 and R4. Molecules governed by fast chemistry (e.g. $n\text{-C}_7\text{H}_{16}$) and short-lived radicals (e.g. O and H) exhibit substantial differences between filtered and unfiltered species concentrations caused by effectively large time scale after filtering. In contrast, the spatial distribution of stable molecules such as CO_2 is sufficiently large scale to be largely unaltered by filtering, with correspondingly minimally changed chemical time scales. As a result, the filtered concentrations of species with small chemical time scales can be significantly higher where the unfiltered concentrations are originally small, as shown for $n\text{-C}_7\text{H}_{16}$, O, and H in Figure 6.

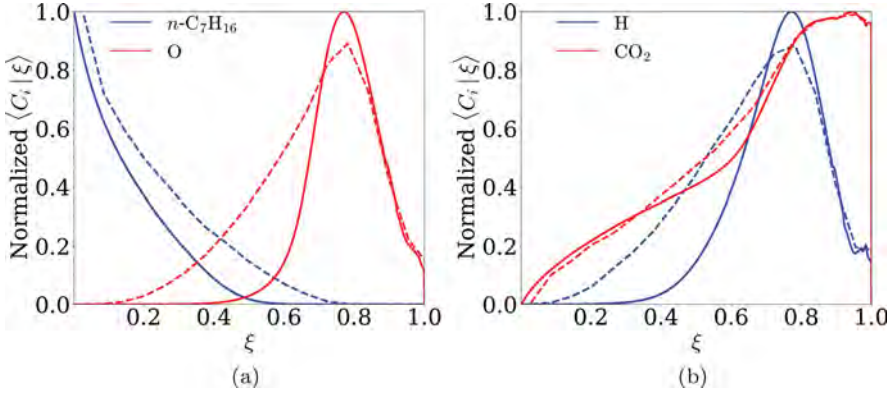


Figure 6. Conditional means of unfiltered (solid lines) and filtered (dashed lines) reactant concentrations over unfiltered and filtered progress variable in the low- Ka flame for reactions R3 and R4 in Table 2. (a) Reactants in R3 and (b) Reactants in R4. (Color online)

Table 3. Reactant chemical time scales and classification for R1–R4.

Species	$\tau(\mu s)$	Classification
$n\text{-C}_7\text{H}_{16}$	0.14	Fast
H	0.46	Fast
O	1.59	Fast
OH	1.69	Fast
CH_2O	1.89	Fast
H_2	39.3	Slow
CO_2	1300	Slow

This makes $\overline{C_i C_j}$ substantially over-predicted by $\overline{C_i} \overline{C_j}$ at these locations, for instance in R1 and R3.

To generalise this analysis to all reactions, the global chemical time scales defined by $\tau_j = \max(\langle |\dot{\omega}_j| / C_j \mid \xi \rangle^{-1})$ for species j are computed for all species in the current chemical model using the unfiltered DNS results for both flames, where $\dot{\omega}$ denotes the species molar production/consumption rate per unit volume. It is found that the chemical time scales are only slightly different at the two Ka , so they are shown in Table 3 only for reactants in R1–R5 for the low- Ka case. Examining the distribution of species chemical time scales τ_j , the 35 species are separated into two clear groups according to $\tau_j \gtrless 10 \mu s$. We refer to $\tau_j \geq 10 \mu s$ as ‘slow’ and $\tau_j < 10 \mu s$ as ‘fast’, respectively. As a result of this classification, stable molecules in the current chemical model such as N_2 , H_2 , H_2O , O_2 , CO , CO_2 are ‘slow’, while the remaining radicals and intermediate species are characterised as ‘fast’.

As such, the disparity in estimating the filtered law of mass action is typically low for reactions between slow species or between fast and slow species, such as R2 and R4. On the other hand, the disparity is generally high for reactions between fast species, such as R1 and R3. In addition to chemical time scales, the disparity for a given reaction also depends on reactant locations within the flame structure. For instance, in reaction R3 where the largest disparity is observed (Figure 5), unfiltered $n\text{-C}_7\text{H}_{16}$ and O are present over ranges of very low and high ξ , respectively, with only a slight overlap between $0.4 \lesssim \xi \lesssim 0.6$,

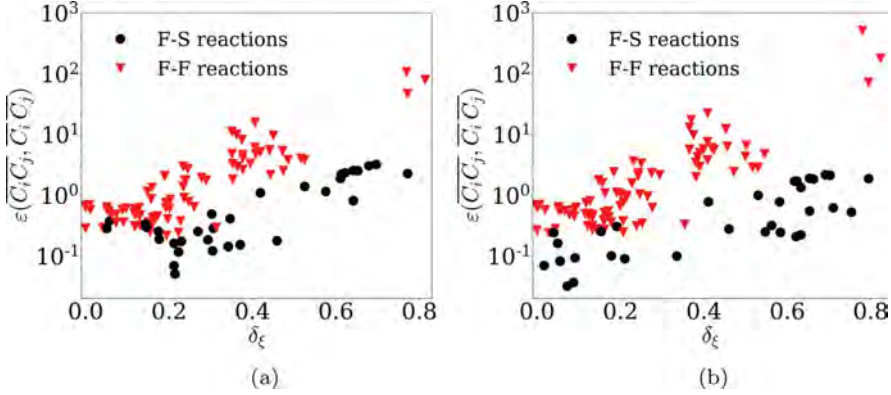


Figure 7. Difference metric ε evaluated between $\overline{C_i C_j}$ and $\overline{C_i} \overline{C_j}$ for all bi-molecular reactions in both flames. (a) Low- Ka case and (b) High- Ka case. (Color online)

whereas the overlap between the two filtered concentrations is broader ($0.2 \lesssim \tilde{\xi} \lesssim 0.7$ in Figure 6). In contrast, the overlap between reactants collocated within the flame structure is likely much less affected by filtering.

Combining the effects of chemical time scales and species formation location, the difference metric, ε , introduced in Equation (4) is computed between $\overline{C_i C_j}$ and $\overline{C_i} \overline{C_j}$ for all bi-molecular reactions. These results are shown in Figure 7 as a function of the difference in reactant formation locations, defined as the difference in the progress variable values where the conditional means of the unfiltered reactants peak, $\delta_\xi = |\xi_{\max,i} - \xi_{\max,j}|$. The reactions are segregated into Fast-Fast (F-F) and Fast-Slow (F-S) according to the reactant chemical time scales. The current chemical model has no S-S reaction. For both flames, F-F reactions typically have larger difference metrics compared to F-S reactions at fixed δ_ξ , as expected. Furthermore, the difference metric increases with increasing δ_ξ for reactions involving reactants formed at significantly different locations, $\delta_\xi > 0.25$. Reactions with $\delta_\xi \lesssim 0.25$ show higher sensitivity to reactant chemical time scales than to δ_ξ . The difference metrics ε computed in the high- Ka flame are shown to be only marginally higher than those in the low- Ka flame for most reactions.

3.4. Disparity in estimating the filtered chemical reaction rates

The disparities in estimating the rate constants and law of mass action are combined in Figure 8, where the conditional mean profiles of the filtered reaction rates $\overline{w}(T, C)$ and the estimated values using filtered thermochemical quantities $w(\tilde{T}, \tilde{C})$ over the Favre-filtered progress variable $\tilde{\xi}$ are compared for reactions R1, R4, and R5 to represent the different reaction types and reactant characteristics. For each reaction the conditional mean profiles are normalised by the maximum value of $\langle w(\tilde{T}, \tilde{C}) | \tilde{\xi} \rangle$. Note that the filtered reaction rate for R5 is artificially amplified in Figure 8 for ease of comparison.

For R1, $\langle \overline{w}(T, C) | \tilde{\xi} \rangle$ is over-predicted by $\langle w(\tilde{T}, \tilde{C}) | \tilde{\xi} \rangle$ by approximately 4 times over $0.1 \lesssim \tilde{\xi} \lesssim 0.8$, which mainly results from the disparity in estimating the filtered law of mass action (Figure 5) since it is a F-F reaction with low E_a . R3 is of the same reaction type, but the difference in reactant formation locations is much larger than R1, which results in a much more severe over-prediction of $\langle \overline{w}(T, C) | \tilde{\xi} \rangle$ by $\langle w(\tilde{T}, \tilde{C}) | \tilde{\xi} \rangle$ (not shown). R4 is a F-S reaction with relatively high E_a , with severely under-predicted filtered rate constant

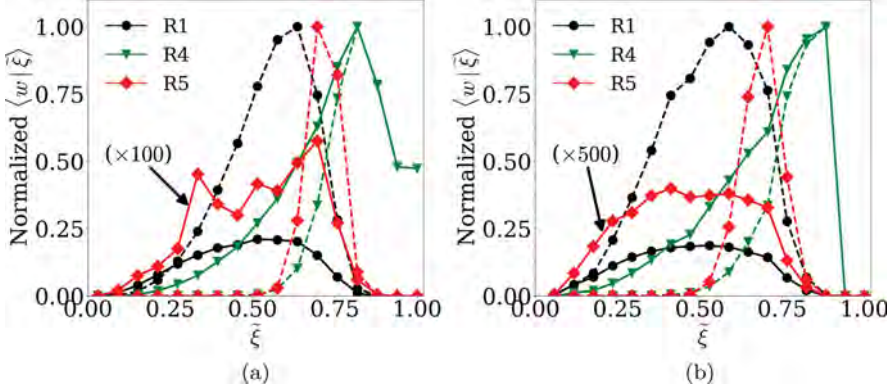


Figure 8. Conditional mean profiles of $\langle \bar{w}(T, C) | \tilde{\xi} \rangle$ (solid lines) and $\langle w(\tilde{T}, \bar{C}) | \tilde{\xi} \rangle$ (dashed lines), normalised by the maximum value of $\langle w(\tilde{T}, \bar{C}) | \tilde{\xi} \rangle$, for R1, R4, and R5 in Table 2. $\langle \bar{w}(T, C) | \tilde{\xi} \rangle$ for R5 is artificially amplified for comparison purposes only. (a) Low-Ka case and (b) High-Ka case. (Color online)

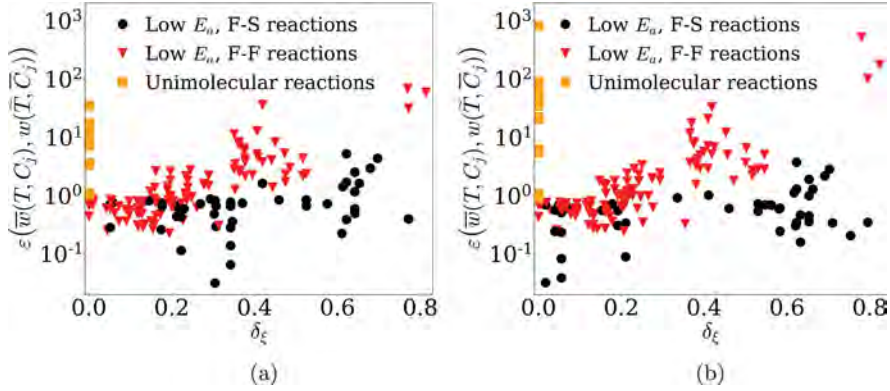


Figure 9. Difference metric ε evaluated between $\bar{w}(T, C_i)$ and $w(\tilde{T}, \bar{C})$ for all reactions in both flames. (a) Low-Ka case and (b) High-Ka case. (Color online)

(Figure 3) and moderately over-predicted law of mass action (Figure 5) over $0.2 \lesssim \tilde{\xi} \lesssim 0.6$. The combined effects lead to severely under-predicted $\langle \bar{w}(T, C) | \tilde{\xi} \rangle$ by $\langle w(\tilde{T}, \bar{C}) | \tilde{\xi} \rangle$ (more than 2 orders of magnitude) over this range of $\tilde{\xi}$ (due to high E_a). R2 is of the same reaction type, but with much lower E_a , for which the under-prediction is only about 50% (not shown). R5 is a uni-molecular fuel decomposition reaction with very high E_a , and the large disparity in estimating its rate constant is shown to occur over $\tilde{\xi} \lesssim 0.5$ (Figure 3), where fuel is present. This results in an under-prediction of the filtered reaction rate by more than 2 orders of magnitude over $\tilde{\xi} \lesssim 0.5$ and an over-prediction by more than 2 orders of magnitude over $0.5 \lesssim \tilde{\xi} \lesssim 0.8$ due to filtering of the fuel concentration (Figure 6(a)). The results are qualitatively the same between the low and high Ka flames.

This analysis is extended to all reactions by computing the difference metric ε between $\bar{w}(T, C)$ and $w(\tilde{T}, \bar{C})$ using Equation (4), as shown in Figure 9. To better classify the reactions based on these disparities, all reactions are segregated into ‘Low- E_a ’ reactions with $E_a \leq 200$ kJ/mol or, otherwise, ‘High- E_a ’ reactions.

This is in addition to the previous designations of ‘Fast-Fast’ (F-F) and ‘Fast-Slow’ (F-S) reactions.

As such, all bi-molecular reactions in the current chemical kinetic model are classified as ‘Low- E_a ’ reactions and only uni-molecular decomposition reactions are qualified as ‘High- E_a ’ reactions. The results indicate that for bi-molecular reactions the disparity in estimating the filtered chemical reaction rates mostly results from the disparity in estimating the filtered law of mass action (Figure 7) due to their relatively low E_a values, especially for ‘F-F’ reactions. The evaluated difference metrics can be as high as 10,000% for these reactions. On the other hand, ‘F-S’ reactions show much higher sensitivity to E_a , but the difference metrics for these reactions are generally quite low ($10\% < \varepsilon < 100\%$). The difference metrics for high- E_a uni-molecular decomposition reactions are generally very high ($100\% < \varepsilon < 100,000\%$), especially in the high- Ka flame. This is due to the combined effects of severely under-estimated rate constants at low $\tilde{\xi}$ and severely over-estimated law of mass action at high $\tilde{\xi}$. Comparing the two flames, although higher Ka generally leads to higher difference metrics for all reactions, the increase is only significant for uni-molecular reactions.

To summarise and generalise our results, bi-molecular elemental reactions typically involve at least one reactant with small chemical time scale, which makes the disparity in estimating the filtered chemical reaction rates generally high for these reactions. Uni-molecular elemental reactions are typically associated with large E_a , which makes the disparity even higher for these reactions. The disparity in estimating the filtered chemical reaction rates would be low only for bi-molecular reactions between reactants with large chemical time scales and low E_a , however, such reactions are rare since reactions between stable molecules typically require very large E_a .

3.5. Variation of the disparity in estimating the filtered chemical reaction rates with filter width

The disparities between the filtered chemical reaction rates and those constructed using the filtered thermochemical quantities are computed for two additional filter widths (in addition to $\Delta = 2l_F$) in each flame simulation to analyse the variation of the disparities with filter width. These filter widths were selected given that the filter size used in practical LES is generally insufficient to resolve the laminar flame thickness (typically with $\Delta \geq l_F$ [1]) to avoid prohibitively high computational cost. Furthermore, the LES grid should be designed to resolve well all integral length scales ($\Delta < l_0/2$). Based on these considerations, filter widths of $1.5l_F$ and $3l_F$ are selected for the low- Ka case, and filter widths of l_F and $1.5l_F$ are selected for the high- Ka case.

The difference metric $\varepsilon(\bar{w}(T, C_j), w(\tilde{T}, \bar{C}_j))$, computed with Equation (4), is plotted as a function of filter width for reactions R1–R5 in Figure 10. As expected, $\varepsilon(\bar{w}(T, C_j), w(\tilde{T}, \bar{C}_j))$ decreases monotonically with decreasing filter width for all reactions. The drop in $\varepsilon(\bar{w}(T, C_j), w(\tilde{T}, \bar{C}_j))$ is more rapid at the smaller filter widths, for instance between $\Delta = 1.5l_F$ and $\Delta = 2l_F$ in comparison with between $\Delta = 2l_F$ and $\Delta = 3l_F$ for the low- Ka case. However, within the range of filter widths considered there is no threshold filter size below which the disparities drop rapidly. More importantly, the order of disparities stays unchanged across different filter widths for the five representative reactions considered, with reaction R2 having the smallest difference metric followed by R4, R1, R5 and R3, consistent with the results shown for $\Delta = 2l_F$ in Section 3.4.

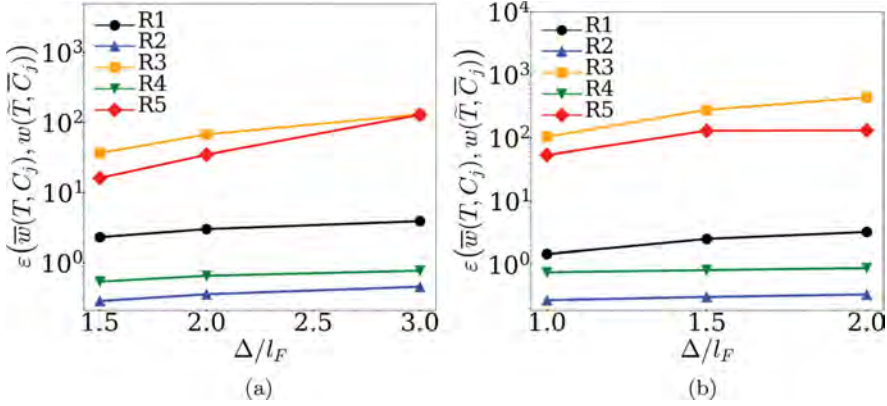


Figure 10. Variation of the difference metric ε evaluated between $\bar{w}(T, C_i)$ and $w(\tilde{T}, \bar{C})$ with filter width Δ for reactions R1–R5. (a) Low- Ka case and (b) High- Ka case. (Color online)

4. Conclusions

An *a priori* assessment of the disparities in estimating the filtered chemical reaction rates in Large-Eddy Simulations (LES) was performed using Direct Numerical Simulations (DNS) of turbulent premixed flames at the same turbulent Reynolds number but different Karlovitz numbers. The sensitivity of these disparities to reaction types, species characteristics, and flame conditions was systematically investigated. The sensitivity to reaction type was examined by quantifying the disparity in estimating the filtered reaction rate constant. The sensitivity to species characteristics was examined by quantifying the disparity in estimating the filtered law of mass action. The sensitivity to flame conditions was examined by contrasting the results obtained from both simulations. For both flames, it was found that the rate constants estimated using LES-filtered temperature severely under-predict the LES-filtered rate constants, especially for reactions with high activation energies, E_a . The law of mass action estimated using LES-filtered species concentrations severely over-predicts the LES-filtered values, especially for reactions involving reactants with shorter chemical time scales present at different locations within the turbulent flame structure. Combining the two effects, it was found that bi-molecular reactions typically involve at least one reactant with small chemical time scale, which makes the disparity in estimating the filtered chemical reaction rates generally high for these reactions. Uni-molecular reactions are typically associated with large E_a , which makes the disparity even higher. The disparity in estimating the filtered chemical reaction rates would be low only for bi-molecular reactions between reactants with large chemical time scales, typically stable molecules, and with low E_a , however, such reactions are rare since reactions between stable molecules typically require very large E_a . These results were found independent of filter width.

Disclosure statement

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Appendix E

An *a priori* Analysis of the Structure of Local Subfilter-Scale Species Surrounding Flame Fronts using Direct Numerical Simulation of Turbulent Premixed Flames

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An *a priori* analysis of subfilter-scale (SFS) species structure important to estimate chemical reaction rates in large-eddy simulation (LES) is performed using direct numerical simulation (DNS) of a turbulent premixed flame at a turbulence Reynolds number $Re_0 = 329$ and Karlovitz number $Ka_0 = 7.23$ with semi-detailed finite-rate chemistry. Differences between the complete chemical reaction rates extracted from DNS and those estimated from LES-filtered variables are quantified. The spatial distributions of these differences are found to be localized in regions surrounding the flame front for representative reactions. Within these regions, variations in the localization relative to the flame, scale, and magnitude of the SFS species concentrations are quantified and mean SFS structure is determined. SFS species structure is found in two groupings: “single-banded” structure characterized by one distinct peak, and “double-banded” structure characterized by two peaks of opposite signs. Species that are produced and consumed within the flame such as CH_2O and HCO are observed to have single-banded structure and species displaying a frontal behavior such as $\text{n-C}_7\text{H}_{16}$ and OH are found to have double-banded structure, on average. The local SFS structure surrounding the flame is impacted by neighboring flame-flame interactions as well as by variations in flame curvature. The impacts of the flame-flame interactions are strong when the SFS species structure has “large” length scales with concentration peaks significantly displaced from the flame front. Curvature effects are shown to be strong in high curvature regions of the flame.

I. INTRODUCTION

The large-eddy simulation (LES) approach has become an important tool for engineering analysis, design and optimization of combustion devices¹. In a properly designed LES, the larger-scale, energy-dominant flow variations are resolved by the effective numerical grid, while the impacts of the unresolved flow and chemical scales on the evolution of the resolved-scales are modeled. Spatial filtering from the grid and implicit coarsening separates an instantaneous field β into resolved-scale (RS) variations β^r with scales larger than the filter size Δ , and subfilter-scale (SFS) variations $\beta^s = \beta - \beta^r$ that are unresolvable and unavailable during LES. The governing equations are solved for the RS quantities, while closure models are needed to describe the effects of SFS quantities at the resolved-scale level through scale-scale interactions, for instance in advective and chemical nonlinearities^{2,3}.

Past research has focussed on the LES modeling of chemical reaction rates using different turbulent combustion models. Examples include low-dimensional manifold-based methods⁴⁻⁶, thickened flame models⁷, transported probability density function (PDF) models^{8,9}, approximate de-convolution methods^{10,11}, conditional moment closure models¹², and linear eddy models^{13,14}. These models are needed because the subfilter-scale variations in thermal energy and species concentrations unavailable in LES can strongly impact the evolution of resolved chemical reaction rates, and therefore the evolution of LES-filtered scalar concentrations and heat release that strongly contributes to the evolution of thermal energy in chemically-reacting turbulent systems¹⁵.

The structure of such small-scale scalar variations has been explored in previous studies with an aim to improve existing models for turbulent combustion. In some early works, Wang and Peters^{16,17} quantified the distribution of scalar structure defined by dissipation elements partitioned by “gradient trajectories” at length scales smaller than the integral scales. They developed a model for the statistical distribution of scalars based on the length scales and scalar difference across these elements. This method was later applied to analyze scalar structure in turbulent premixed and non-premixed jet flames^{18,19}. Other research has focussed on analysis of the correlations between specific flame curvatures and small-scale scalar structure.

For example, combined studies of local flow topology and curvatures of iso-scalar surfaces^{20,21} have found that focal topologies, i.e. topologies characterized by one real and a pair of complex-conjugate eigenvalues in velocity gradient tensor, are dominant in regions closer to unburnt reactants, while nodal topologies characterized by three real eigenvalues of velocity gradient tensor are prevalent near hot products. Small-scale scalar structure has been studied in turbulent flames for different flame geometries (characterized in terms of local curvatures of scalar iso-surfaces) in relationship to volumetric dilatation rate and tangential and normal strain rate^{20,21}. Other studies have investigated scalar structures conditioned on local scalar dissipation rate for different dilatation levels and different flame curvatures to identify the relationships between the local scalar structure and scalar dissipation rate²². However, the structures underlying the scalar variations at unresolved scales within the LES modeling framework, and the extent to which SFS scalar structures are related to variations in flame geometry, have not been studied. Knowledge potentially useful in structure-based SFS modeling strategies, specifically, the understanding of structure common to the variations of

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SFS species concentrations in regions where they impact the resolved contributions to chemical reaction rates in LES is useful for improved modeling approximations of SFS contributions to chemical reaction rates in LES of premixed turbulent combustion.

To this end, our main objective is to quantify and characterize the differences in the structure of SFS species concentrations in relation to species type, flame curvature and flame-flame interactions using DNS-based *a priori* analysis. We first identify reaction-specific “zones of influence” defined as the region localized to the flame-front where the incorporation of SFS species concentrations has a strong impact on the estimation of chemical reaction rates in LES. Within these zones of influence, we classify SFS species structure averaged over the flame-sheet into distinct groupings and quantify the scale, magnitude and location of SFS species concentrations. For each species, we then investigate the extent of deviations from the mean SFS species structure due to flame-flame interactions and variations in the flame curvature. The main intent of this work is not to develop new models to improve the predictive capabilities of LES, but rather to analyze specific characteristics of SFS species structure for applications to future SFS model developments in LES.

II. DNS OF A TURBULENT PREMIXED FLAME

A. Flow Configuration and DNS Setup

The analysis is performed using the DNS dataset of an *n*-heptane/air turbulent premixed flame (referred to there as the “low-*Ka*” case) from a previous study¹⁵. Here, only a brief summary of the DNS setup and numerical methods is presented. The configuration of the simulation is that of a statistically-planar flame, with inflow/outflow boundary conditions in the stream-wise direction and periodic boundary conditions in the span-wise directions as shown in Fig. 1. A fully-premixed, slightly lean *n*-C₇H₁₆/air mixture is injected at the inflow plane with unburnt temperature $T^u = 300$ K at 1 atm pressure and equivalence ratio $\phi = 0.95$. Under these conditions, the laminar flame thickness and burning velocity were computed to be $l_F = 3.15 \times 10^{-4}$ m and $s_L = 0.39$ m/s, respectively, using the chemical model described in Section II B. Homogeneous isotropic turbulence (HIT) is injected at the inflow plane that was generated from a separate incompressible 3D turbulence simulation at stationary state with spectral forcing²³ with correlation-based integral scale $L_{iso} \equiv 1/3 \sum_{\alpha=1}^3 l_{\alpha\alpha,\alpha} = 7.05 l_F$.

To avoid any potential artifacts from turbulence forcing, no forcing is imposed in the turbulent flame simulation and turbulence is allowed to decay freely as it interacts with the flame downstream. The domain is discretized using 900 points in the stream-wise and 500 points in the span-wise directions, with a resolution of at least 10 points across the laminar flame. The grid is non-uniformly stretched downstream of the flame in the stream-wise direction. The integral length scale at the intended mean flame location about $4.3 L_{iso}$ downstream to

the inflow plane is given by:

$$l_0 = \frac{1}{2} (l_{22,2} + l_{33,3}), \quad (1)$$

where $l_{22,2}$ and $l_{33,3}$ are the integral length scales based on velocity component correlations in the transverse directions. $l_{11,1}$ is not included in the above equation because the stream-wise direction is inhomogeneous. The Kolmogorov length scales at this location are computed as:

$$\eta_0 \equiv \left(\frac{\nu^3}{\langle \epsilon \rangle_0} \right)^{1/4} = 0.13 l_F, \quad (2)$$

where ν and ϵ denote the viscosity of the unburned reactant mixture and the turbulence dissipation rate respectively. The RMS turbulent velocity fluctuations at the intended mean flame location are computed as:

$$u_0 \equiv \sqrt{(\langle u_2^2 \rangle_0 + \langle u_3^2 \rangle_0)/2} = 3.38 s_L. \quad (3)$$

In the Equations (1)-(3) above, the subscript 0 implies that the ensemble averages $\langle \cdot \rangle_0$ are calculated as averages over the transverse plane and in time for about 30 eddy-turnover times (l_0/u_0) at the average location of the flame front in a separate DNS of non-reacting, incompressible decaying turbulence with the same inflow and computational domain as the actual flame simulation. At this location, the turbulent Reynolds number is computed as $Re_0 = u_0 l_0 / \nu = 329$. The Karlovitz number²⁴ is computed as

$$Ka_0 \equiv \frac{t_F}{t_\eta} = \frac{l_F \nu}{s_L \eta_0^2} = \left(\frac{\nu}{s_L l_F} \right) \left(\frac{l_F}{\eta_0} \right)^2 \approx 7.23, \quad (4)$$

where t_F and t_η are the flame and Kolmogorov time-scales. This value of Re_0 is chosen so that scale ratio l_0/l_F is at least 10 at the expected flame location. With these specified conditions, the flame is expected to fall in the thin reaction zones regime.

B. Computational Methodology

A semi-detailed chemical kinetic model for *n*-heptane/air laminar premixed flame containing 35 species and 217 reactions²⁵ (forward and backward reactions counted separately) was used. This mechanism is reduced from the original model with 149 species and 1651 reactions²⁶. Complete details of the chemical model and its validation are provided in Lapointe et al.²⁵ for DNS of high-*Ka* turbulent premixed flames. The turbulent flame simulations were performed using the NGA code²⁷, where the unsteady Navier-Stokes equations with the low Mach number approximation and the transport equations for temperature and species mass fractions are solved simultaneously. The scalar equations are discretised using the BQUICK scheme, which ensures that the physical bounds of appropriate quantities are numerically preserved without adding significant artificial diffusion²⁸. A recently developed computationally-efficient, semi-implicit, iterative method is

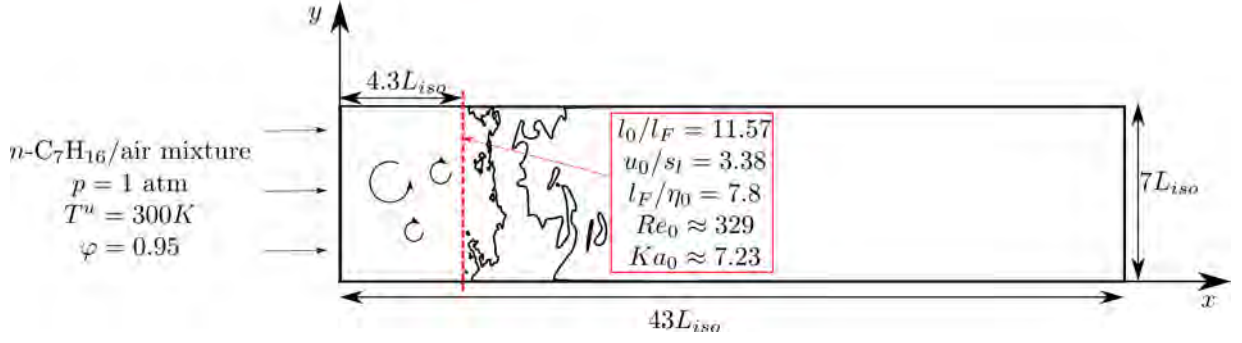


FIG. 1: Two dimensional schematic of the domain with simulation configuration details.

used for the time-integration of chemical source terms²⁹. This method has been shown to be free of lagging errors, is as efficient as explicit time integration, and is capable of maintaining stability with relatively large time-steps³⁰.

III. FRAMEWORK FOR ANALYSIS OF THE DNS DATASET

A. Filtering of the DNS Dataset

To represent the filtered LES fields, the DNS dataset is filtered *a priori* using a top-hat box spatial filter with a fixed filter width. In LES, the effective filter width should resolve a large fraction of the momentum and temperature variance, where we interpret “large” as at least 80%. Considering these criteria, we find that a filter width of $\Delta = 2l_F = 630\mu\text{m}$ resolves $> 85\%$ of the variance in momentum and enthalpy per unit volume, $\rho\mathbf{u}$ and ρh , identified by systematically varying the filter width down to the DNS grid resolution. The current filter width has been used in other LES for premixed turbulent combustion^{31,32}. With the application of the filtering procedure, any variable β is decomposed into resolved-scale (RS) β^r and subfilter-scale (SFS) $\beta^s = \beta - \beta^r$ quantities. These RS and SFS quantities are computed at each spatial location on the DNS grid. In Fig. 2, we show RS vs full temperature T , and species concentrations $\theta = \rho Y$ for fuel ($\theta_{n\text{-C}_7\text{H}_{16}}$) and the intermediate species formaldehyde ($\theta_{\text{CH}_2\text{O}}$) on a planar slice extracted from the three-dimensional domain. Figure 2 shows that the filtering process thickens the flame structure and smears the high gradients to filter scale. This is especially apparent with species formed within the flame such as CH_2O .

B. Classification of Species and Reactions

Based on their variances at resolved scales, we classify all species into “R” species with $\geq 85\%$ of the total variance in the resolved scales, “S” species with $\leq 20\%$ of the total variance in the resolved scales, and “M” species with variance between 85% and 20% in the resolved scales. Note that these variances are evaluated after removing the global mean of species concentration. The full list of species and their resolved-scale variances may be found in the supplementary material. Based

on this species classification, we identify six representative reactions and participating species listed in Table I for analysis. These include different reactions between species belonging to each of these three classes. These reactions are also chosen to incorporate a range of different chemical kinetic rate parameters, including both positive and negative values of the temperature exponent, n , and activation energy, E_a . These kinetic parameters indicate the extent of the non-linear dependence of the reaction rate constant $k(T)$ on temperature expressed using the Arrhenius rate law,

$$k(T) = AT^n \exp\left(-\frac{E_a}{RT}\right). \quad (5)$$

Additionally, these reactions are chosen to contain different reaction types such as exothermic (R1) and endothermic (R2) reactions, bi-molecular oxidation reactions (R1, R2) and uni-molecular decomposition reactions (R4). These reactions include stable molecules and free radicals as well as species among the top ten contributors to average heat release rate ($n\text{-C}_7\text{H}_{16}$, CO_2 , OH , H). This set of reactions (R1-R5) has also been used in a previous study of similar nature¹⁵.

TABLE I: Representative reactions and their corresponding kinetic rate parameters. Variance classification of reactants in each reaction are indicated in parenthesis with (R) and (S) denoting species with resolved scale variance $\geq 85\%$ and $\leq 20\%$ respectively, and with (M) to denote species with intermediate range of RS variances.

	n	E_a (kJ/mol)	Reaction
R1	1.2	-1.87	$\text{CH}_2\text{O}(\text{M}) + \text{OH}(\text{R}) \rightarrow \text{HCO} + \text{H}_2\text{O}$
R2	1.5	14.4	$\text{OH}(\text{R}) + \text{H}_2(\text{M}) \rightarrow \text{H} + \text{H}_2\text{O}$
R3	2.8	9.46	$n\text{-C}_7\text{H}_{16}(\text{R}) + \text{O}(\text{R}) \rightarrow 2\text{-C}_7\text{H}_{15} + \text{OH}$
R4	-1.1	106	$\text{H}(\text{R}) + \text{CO}_2(\text{R}) \rightarrow \text{OH} + \text{CO}$
R5	-17.6	504	$n\text{-C}_7\text{H}_{16}(\text{R}) \rightarrow 1\text{-C}_5\text{H}_{11} + \text{C}_2\text{H}_5$
R6	2.15	76.87	$\text{H}_2(\text{M}) + \text{HCO}(\text{S}) \rightarrow \text{H} + \text{CH}_2\text{O}$

C. Normal Line Representation on Discretized Flame-sheet

For the quantification of the SFS structure surrounding the flame, we define the flame sheet as the $C^r = 0.5$ iso-surface of

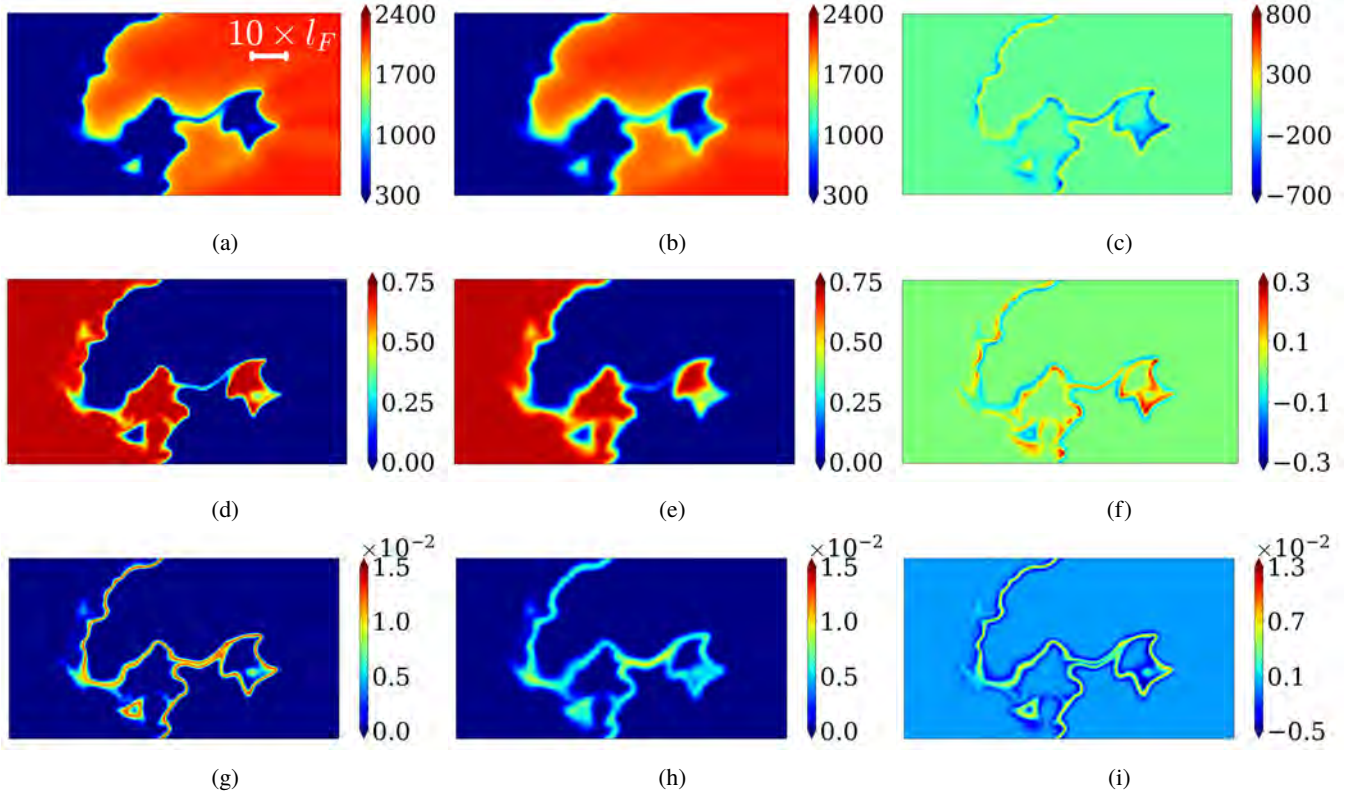


FIG. 2: Original¹⁵, RS and SFS variables: (a) T (K); (b) T^r (K); (c) T^s (K); (d) $\theta_{n-C_7H_{16}}$ (mol/m³); (e) $\theta_{n-C_7H_{16}}^r$ (mol/m³); (f) $\theta_{n-C_7H_{16}}^s$ (mol/m³); (g) θ_{CH_2O} (mol/m³); (h) $\theta_{CH_2O}^r$ (mol/m³), and (i) $\theta_{CH_2O}^s$ (mol/m³). A length scale corresponding to $10 \times l_F$ is shown in panel (a). Images in panels (a), (d) and (g) are reproduced from Shah et al.¹⁵

a temperature-based resolved-scale progress variable,

$$C^r = \left(\frac{T^r - T^u}{T^b - T^u} \right), \quad (6)$$

similar to previous studies³³. Here, T^u and T^b are the temperatures of the unburned fuel and the burned products respectively. The level $C^r = 0.5$ is chosen to approximate the peak heat release within the flame.

To identify the common structure underlying the spatial distribution of subfilter-scale scalar quantities, we numerically construct curvilinear normal lines from the flame-sheet and analyze the quantities of interest along individual lines. In addition, a separate set of straight normal lines is constructed for the analysis of flame-flame interactions on species SFS structure detailed in Section IV C 1, as the curvilinear normal lines cannot intersect a nearby flame-sheet.

An example of a curvilinear normal line is shown in Fig. 3. To construct these lines, first the flame-sheet iso-surface $C^r = 0.5$ is numerically realized as a set of discrete locations $\mathbf{y}_j$ ($j = 1, 2, \dots, N$) which satisfy $C^r(\mathbf{y}_j) = 0.5$. This set of points is obtained by applying the well-known marching cubes algorithm³⁴ to the filtered progress variable, which partitions the flame-sheet into sets of connected triangular elements shown in Fig. 3 with their nodes representing the discretized locations $\mathbf{y}_j$ on the flame-sheet. The application of this algo-

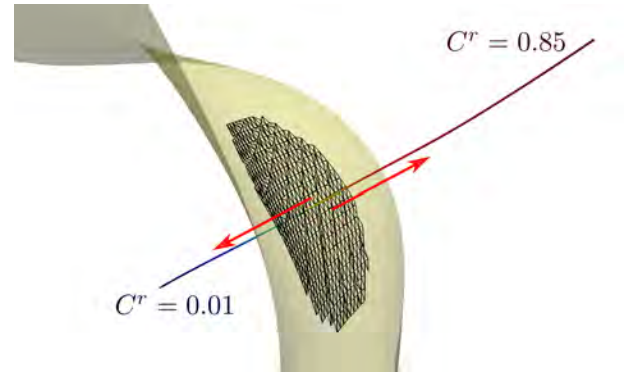


FIG. 3: Illustration of a curvilinear normal line originating from a node on the discretized mesh (black) on the flame-sheet ($C^r = 0.5$) shown in yellow. The normal line is colored based on C^r in the range 0.01 (blue) - 0.85 (red). Note that only a part of the flame-sheet is shown for illustration.

algorithm results in a discretized flame-sheet with $N = 1.78 \times 10^6$ nodes which are non-uniformly distributed with the distance between any two adjacent (connected) nodes not exceeding $0.17l_F$.

The curvilinear normal lines are constructed starting from

the j^{th} node $\mathbf{y}_j$ on the discretized flame front, by following the normal direction on both sides of the flame-sheet - one facing the unburned reactants and the other facing the burned products. Points on each normal line are uniquely represented as a function of the signed curvilinear distance n from the starting node on the flame sheet ($n = 0$), such that $n < 0$ and $n > 0$ correspond to distances on the unburned reactant side and burned product side respectively. Numerically, these points $\mathbf{x}(n)$ are obtained for the j^{th} normal line by solving the following equation,

$$\frac{d\mathbf{x}}{dn} = \frac{\nabla C^r(\mathbf{x})}{|\nabla C^r(\mathbf{x})|}, \quad (7)$$

using explicit Euler integration scheme along descending $n < 0$ and ascending $n > 0$ gradient directions, with initial conditions $\mathbf{x}(0) = \mathbf{y}_j$ and a step-size $\Delta n = l_F/20$. Here, the gradients of C^r are computed first on the original mesh using second order accurate central difference scheme and later interpolated linearly to locations $\mathbf{x}$ on the normal line. As such, $C^r(\mathbf{x})$ changes monotonically in each direction along the normal lines and the integration is terminated when $C^r(\mathbf{x}) = 0.01$ is reached for $n < 0$ and $C^r(\mathbf{x}) = 0.85$ is reached for $n > 0$. On these normal lines, the original, resolved-scale and subfilter-scale quantities are expressed a function of n .

IV. RESULTS AND DISCUSSIONS

A. Spatial Distribution of Impacts to the Estimation of Chemical Reaction Rates from the Incorporation of SFS Species Concentrations : Zones of Influence

We analyze the SFS structure of species concentrations in regions where the incorporation of SFS species concentrations is impactful to the estimation of the reaction rate nonlinearities in LES. To identify the spatial extent of these regions, termed ‘‘zones of influence’’, the impact from incorporation of SFS species concentrations in reaction i is computed on the j^{th} normal line as the absolute difference $\delta_{i,j}(n)$ between the complete reaction rate $w_{i,j}(n; T, \theta)$ constructed from the DNS data, including SFS content, and reaction rates constructed from LES-filtered quantities $w_{i,j}(n; T^r, \theta^r)$, normalized by the average reaction rate $w_{i,j,avg}$,

$$\delta_{i,j}(n) = \frac{|w_{i,j}(n; T^r, \theta^r) - w_{i,j}(n; T, \theta)|}{w_{i,j,avg}}. \quad (8)$$

The averaging in $w_{i,j,avg}$ is performed in the interval over which $w_{i,j}(n; T, \theta)$ differs significantly from zero; this is determined separately for each normal line by requiring that $w_{i,j}$ is larger than 10% of the maximum reaction rate along the normal line. We have also performed the analysis presented in this section with a much smaller tolerance level of 1% to confirm that our results are relatively insensitive to the tolerance level within this range (not shown). The reaction rate $w_{i,j}$ is computed for a generic reaction i , $\sum_{r=1}^{N_i} \nu_r' A_r \rightarrow \sum_{r=1}^{N_i} \nu_r'' A_r$,

on the j^{th} normal line using the ‘law of mass action’ as,

$$w_{i,j}(n; T, \theta) = k_i(T(n)) \prod_{r=1}^{N_i} [\theta_r(n)]^{\nu_r'}. \quad (9)$$

In the above reaction, ν_r' and ν_r'' are the stoichiometric coefficients of species r on the reactants and products sides respectively and N_i is the total number of species involved in reaction i . $k_i(T)$ is the temperature-dependent reaction rate constant expressed using the Arrhenius rate law (Eq. (5)).

Example profiles of the chemical reaction rates constructed from full and LES-filtered quantities for reaction R1 are shown for four different normal lines in Fig. 4(a) as a function of n . In general, the reaction rate is expected to be small away from the flame, into the unburned reactants ($n < -4l_F$) and burned products ($n > 4l_F$).

Figure 4(b) shows that the $\delta_{i,j}$ is localized near the flame-sheet approximately between $-2l_F$ and $2l_F$ and up to 2.5 times higher in magnitude than the average reaction rate along each normal line shown in Fig. 4(a). For each normal line, the zone of influence is identified as the interval $[n_{i,j}^l, n_{i,j}^u]$ over which the differences between the chemical reaction rates from DNS data and those constructed from LES-filtered quantities are at least 10% of the average reaction rate (i.e. $\delta_{i,j} > 0.1$). In Fig. 4(b) the limits of the influence zone can be identified for the example normal lines by the intersection of the horizontal dotted line with the individual profiles.

To generalize the quantification of influence zones to the entire flame, the statistical distribution of lower ($n_{i,j}^l$) and upper ($n_{i,j}^u$) limits of the influence zone is obtained over the ensemble of normal lines from the flame-sheet. Figure 4(c) shows the PDF of lower and upper limits of the influence zone interval for reaction R1. This PDF is obtained by weighting the influence zone limits for each normal line with weights proportional to the area A_j of the flame-sheet surrounding the starting node for each normal line. This surface-weighted averaging scheme is used to account for the non-uniform distribution of normal lines from their starting nodes on the flame sheet, similar to previous studies¹⁹.

The distribution of the lower and upper limits of the influence zone in Fig. 4(c) shows that the most probable values are at $n = -0.88l_F$ and $n = 1.52l_F$ respectively, with normal lines from more than 99% of the flame surface having a zone of influence within $-2l_F$ and $2.46l_F$ for reaction R1. These limits along with their corresponding influence zone intervals are quantified relative to l_F for reactions R1-R6 in Table II. These results indicate that the overall influence zone based on the disparities for all reactions are localized within $-3.3l_F$ and $2.46l_F$ from flame-sheet.

In Table II, reactions R1 and R3 are shown to have the lowest and highest overall limits, with influence zone interval sizes exceeding $4l_F$. These are also the reactions associated with larger disparities with the filtered chemical reaction rates as reported in previous work¹⁵. In comparison, reactions R2, R4 and R6 show a smaller zone of influence ($< 3l_F$). Considering the diverse set of reactions and species included in this analysis, the overall influence zone may be expected to be localized within $\pm 4l_F$ from the flame-sheet. In principle, the incorpo-

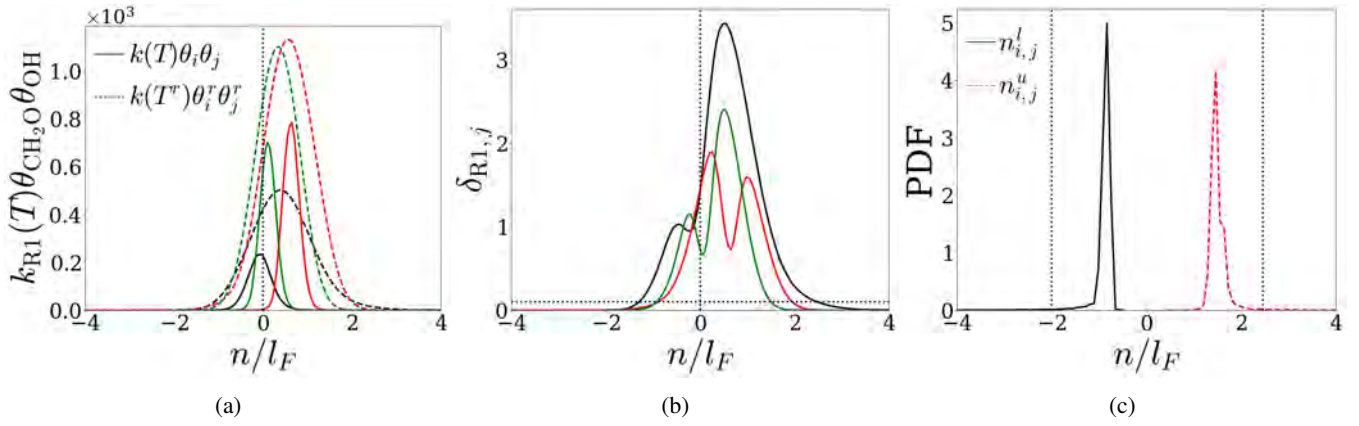


FIG. 4: (a) Reaction rate for reaction R1 along select normal lines computed from full and filtered fields, (b) Disparity between the two reaction rates and (c) Area averaged PDF of lower and upper bounds from entire flame surface. Profile from each normal line is highlighted by its distinct color in (a) and (b). Dotted black line in (b) indicates the 10% level threshold for δ_j . Dotted black lines in (c) indicate zone of influence for R1 over 99% of flame-sheet area.

TABLE II: Zones of influence based on most probable bounds (Zone I) and 99% limits (Zone II) over which $\delta_{i,j}(n) > 0.1$ for reactions R1-R6 in Table I. All quantities are shown relative to l_F .

Reaction	Lower bound (99%)	Most probable lower bound	Most probable upper bound	Upper bound (99%)	Influence Zone I	Influence Zone II
R1	-2.0	-0.88	1.52	2.46	2.40	4.46
R2	-0.5	-0.23	1.16	1.75	1.39	2.25
R3	-3.3	-1.11	1.39	2.20	2.40	5.50
R4	-0.05	-0.18	1.43	2.34	1.61	2.39
R5	-0.6	-0.28	1.44	2.40	1.72	3.00
R6	-0.45	-0.24	1.35	2.30	1.59	2.75

ration of SFS content through LES models confined only to these regions is expected to have an impact on improving LES estimates of chemical reaction rates.

B. Structure of Local SFS Concentrations

1. Overall Characteristics of SFS Quantities

Within the influence zones, the SFS species concentrations θ_i^s are assessed for reactants in R1-R6 by mapping them on normal lines originating from the flame-sheet. The full, resolved-scale and subfilter-scale temperature and concentrations of species in reaction R1 (CH_2O and OH) are shown in Fig. 5 between $-4l_F$ and $4l_F$ relative to the flame-sheet for the same example normal lines shown in Fig. 4. For each of these normal lines, the full temperature and OH concentration exhibit a frontal variation with n , while CH_2O concentration shows a distinct peak near $n = 0$ and diminishes away from the flame for $|n| > 2l_F$.

The temperature increases across the flame near $n = 0$ due to heat release resulting in the frontal variation. The RS temperature approaches the full temperature away from the flame near the unburned reactants ($n < -2$) and burned products ($n > 2$) due to reduced spatial variations from mixture homogeneity in the pre-flame region and reduced turbulence intensity in the

post-flame region. Across the flame, the RS temperature is shown to have a milder front as the filtering process smears out the spatial gradients. As a consequence, T^r over-estimates T roughly for $n \lesssim 0$ and under-estimates T for $n \gtrsim 0$. These differences between T and T^r result in a “double-banded” pattern in the SFS temperature T^s (Fig. 5(d)) characterized by a positive and negative peak on either side of the flame-sheet. A similar pattern is observed for OH where the front-like structure is due to the increase in concentration near the oxidation layer downstream of the reaction zone. As a consequence, $\theta_{OH}^r \geq \theta_{OH}$ for $n < 0$ and $\theta_{OH}^r < \theta_{OH}$ for $n > 0$ resulting in a double-banded pattern for SFS OH concentration in Fig. 5(f).

In the case of an intermediate species produced in the flame such as CH_2O , the concentration peaks near the flame-sheet and diminishes away from the peak location. RS CH_2O concentration appears to have a larger spread compared with the full field due to the smearing out of gradients by the filtering process. The smeared-out profile is shown to underestimate the actual concentration near the peak in Fig. 5(b), while the spread causes the RS concentration to be over-estimated away from the peak location. These differences lead to a “single-banded” pattern in the SFS CH_2O concentration characterized by a positive peak where $\theta_{CH_2O} > \theta_{CH_2O}^r$ and surrounded by negative peaks of smaller magnitude in locations where $\theta_{CH_2O} < \theta_{CH_2O}^r$, with diminishing concentration away from the peaks as shown in Fig. 5(e). Based on these observations, species that are lo-

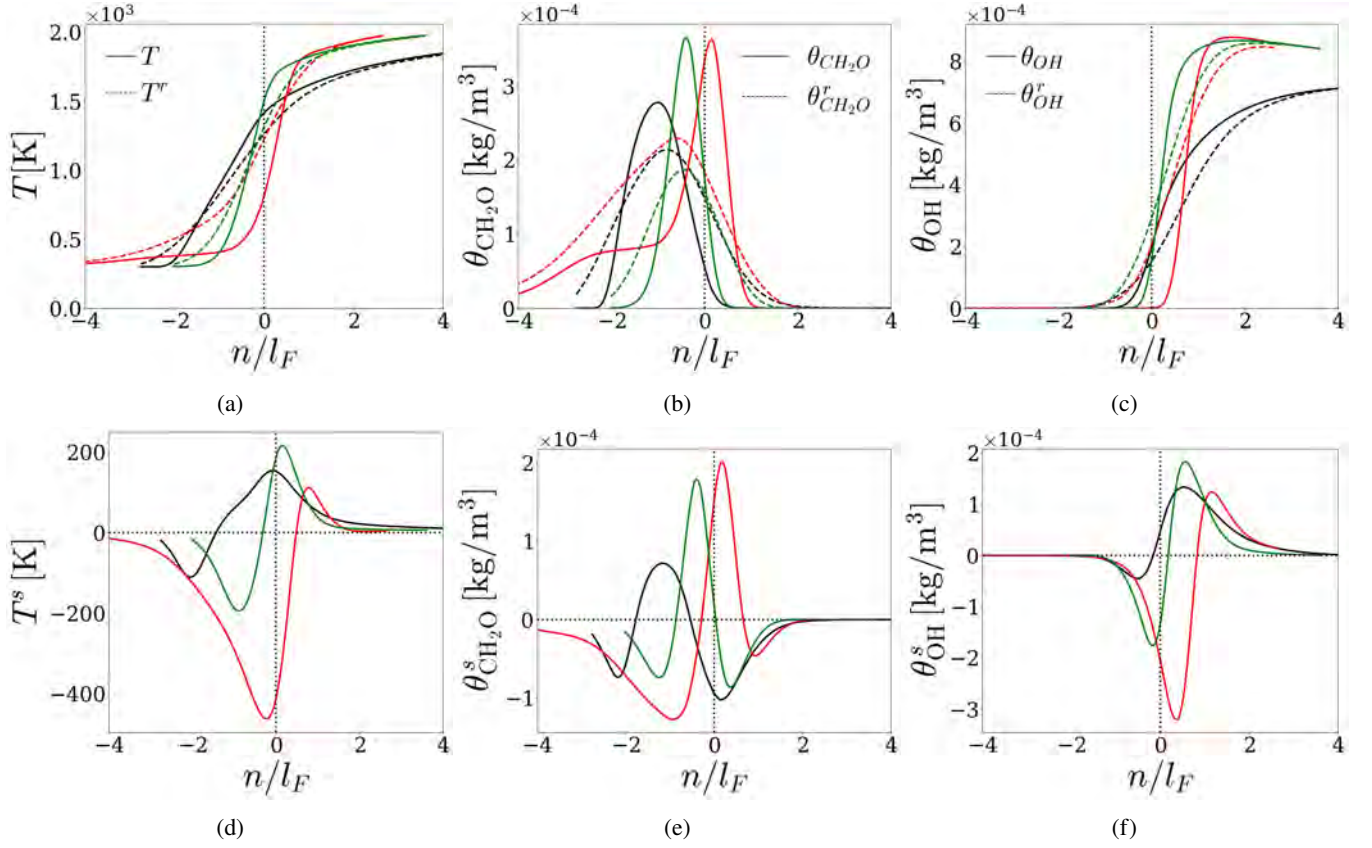


FIG. 5: Full (solid), resolved-scale (dashed) profiles (top row) and corresponding subfilter-scale profiles (bottom row) for Temperature, CH_2O and OH concentration along select normal lines shown in Fig. 4. Profile along each normal line is distinguished by the color assigned to it.

cal to the flame are expected to show single-banded patterns, while species exhibiting a frontal variation are expected to show double-banded patterns in their SFS concentrations.

2. Parameterization of SFS Concentrations on Normal Lines

Although the SFS concentration profiles for each species appear to have a common single or double-banded structure, the SFS profile along each normal line has a distinct characteristic scale, location and magnitude. We systematically define these characteristics and make an assessment of the variability in profiles across all normal lines originating from the flame-sheet. For each normal line, we characterize the location n_p and scale l_θ of the single-banded SFS forms by their peak location and the curvilinear distance along normal line between zero cross-over locations surrounding the peak. The magnitude for single-banded SFS structure is defined as the SFS concentration at the peak location $\theta_i^s(n = n_p)$. In the case of double-banded forms, the location n_p is defined by the zero cross-over point between the negative and positive peaks. The scale l_θ is defined as the curvilinear distance between positive and negative peak locations and the magnitude is defined as the mean of the absolute values of SFS concentration at both peak locations. Figure 6 shows the PDF of location and

scales of the local SFS weighted by the flame-sheet area surrounding starting nodes of each normal line. Three species, CH_2O , HCO , and H_2 are observed to have a single-banded form and the remaining species, OH , H , O , $n\text{-C}_7\text{H}_{16}$, and CO_2 have forms that resemble a double-banded structure. Among the species showing single-banded forms, the most probable peak location for HCO occurs at $n_p = 0.09l_F$ and those for CH_2O and H_2 are at $-0.36l_F$ and $0.15l_F$ respectively. For species showing double-banded forms, the most probable location for fuel $n\text{-C}_7\text{H}_{16}$ occurs at $n_p = -0.58l_F$. Hydrogen radical H has its most probable location near the flame-sheet at $n_p = 0.052l_F$ and the remaining species OH , O and CO_2 have their most probable locations at $0.29l_F$, $0.26l_F$ and $0.37l_F$ respectively. SFS concentration profiles conforming to double-banded forms show a larger spread in locations in Fig. 6(a,c) compared to those showing single-banded forms.

Stable molecules such as $n\text{-C}_7\text{H}_{16}$, CO_2 , CH_2O and H_2 are shown to have highest probability values at larger characteristic length scales $1.05l_F$, $1.67l_F$, $0.89l_F$ and $1.05l_F$ respectively, as these species have a greater tendency to spread out over larger length scales due to their larger chemical time-scales. Initial reactants and final combustion products $n\text{-C}_7\text{H}_{16}$ and CO_2 have a larger variability in scale compared with other species. Radicals such as OH , H , O , and HCO are more likely to have smaller scales as shown in Fig. 6(b,d), due

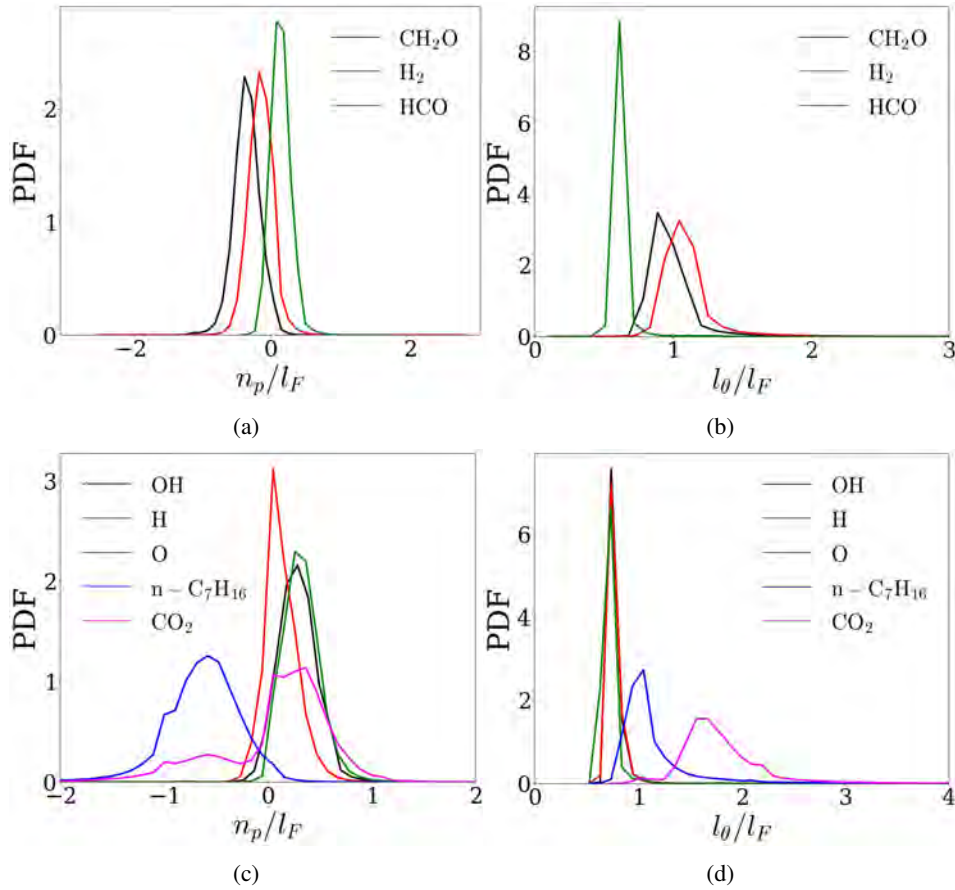


FIG. 6: Variability in location (left) and scale (right) of SFS concentration relative to l_F : single-banded (top) and double-banded (bottom) species.

to their small chemical time scales .

These trends are reflected in the area weighted statistics of parameter distribution reported for each species in Table III. The mean location $\langle n_p \rangle$ for SFS concentration profiles along all normal lines are between $\langle n_p \rangle = \pm 0.65 l_F$ placing them well within the overall influence zone (Section IV A) on average. The mean location is closest to flame-sheet for CO_2 . The standard deviation $\sigma(n_p)$ in n_p is higher for $n\text{-C}_7\text{H}_{16}$ and CO_2 at $0.38 l_F$ and $0.56 l_F$ respectively when compared with remaining species ($0.14 l_F \leq \sigma(n_p) \leq 0.19 l_F$), consistent with the spread shown in Fig. 6(b,d). In terms of the mean scale $\langle l_\theta \rangle$, HCO , OH , H and O have scales smaller than laminar flame thickness ($\langle l_\theta \rangle < l_F$) with HCO being the smallest ($\langle l_\theta \rangle = 0.62 l_F$). These are also the species with smaller standard deviations in scale ($0.07 \leq \sigma(l_\theta) \leq 0.09$) compared with other species in Table III. The mean scales for CO_2 is largest followed by $n\text{-C}_7\text{H}_{16}$. Both species have larger standard deviations in scale $\sigma(l_\theta) > 0.4 l_F$ compared with other species.

Note that the length scales shown in Fig. 6 and Table III are for the SFS species profiles, which are different from the physical length scales of each species (e.g., based on the characteristic velocity and chemical time scales). The length scales for SFS species profiles are affected not only by the physical

TABLE III: Mean ($\langle \cdot \rangle$) and Standard deviations ($\sigma(\cdot)$) of location and scale of SFS concentration for single and double banded forms.

species	$\langle n_p \rangle / l_F$	$\sigma(n_p) / l_F$	$\langle l_\theta \rangle / l_F$	$\sigma(l_\theta) / l_F$
CH_2O	-0.34	0.18	0.98	0.22
H_2	-0.15	0.19	1.09	0.21
HCO	0.13	0.14	0.62	0.07
OH	0.28	0.19	0.76	0.09
H	0.14	0.17	0.75	0.09
O	0.33	0.18	0.71	0.09
$n\text{-C}_7\text{H}_{16}$	-0.64	0.38	1.15	0.43
CO_2	0.07	0.56	1.79	0.41

length scales of the full species profiles, but also by the LES filter width, since it measures the average spread of the SFS profiles. Because of this, species with fast chemistry and low diffusivity, such as $n\text{-C}_7\text{H}_{16}$, may have relatively large SFS length scales, comparable to other species with high diffusivities, such as H_2 .

To summarize, these trends are consistent with the overall chemical characteristics of these species. The mean peak location and scales of CH_2O as described in Table III are expected as CH_2O is present predominantly in the preheat

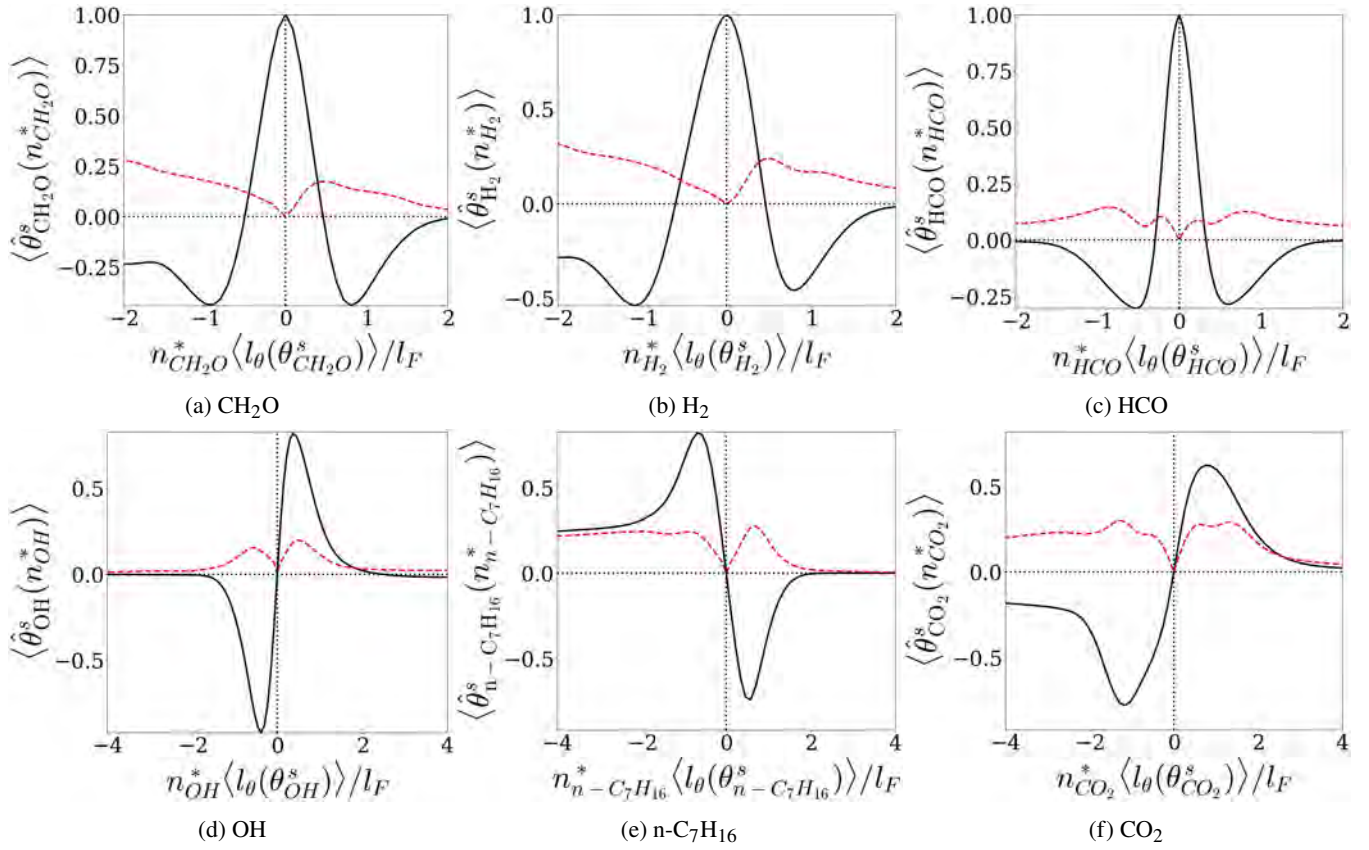


FIG. 7: Mean (black, solid) and standard deviation (red, dashed) in SFS concentration structure across all normal lines weighted by the area assigned to each normal line for single-banded (top) and double-banded (bottom) species.

zone³⁵. HCO being a dominantly SFS variable localized to the reaction zone has smaller mean length scales, and fuel being dominant near unburned reactants has the most negative $\langle n_p \rangle$. On the other hand, species in the oxidation zone such as OH, O have higher positive $\langle n_p \rangle$. As CO₂ is formed near post-flame regions, it has a positive mean location. Stable species with larger chemical time-scales such as n-C₇H₁₆ and CO₂ tend to have larger mean length scales.

3. Basic Forms of Ensemble Averaged SFS Concentration

To obtain the structure of SFS concentrations averaged over the entire flame and quantify the variability in the structure over the entire flame, it is necessary to factor out the variability in the location, scale and magnitude of the local SFS concentrations from each individual normal line. To achieve this, we introduce a linear transformation of the variable n that allows the structure to be compared across different normal lines by aligning the locations to a common origin and scaling the n values by the length scales of individual profiles. This is given by,

$$n_{i,j}^* = \frac{n - n_{p,j}(\theta_i^s)}{l_{\theta,j}(\theta_i^s)}, \quad (10)$$

where $n_{p,j}(\theta_i^s)$ and $l_{\theta,j}(\theta_i^s)$ are the location and scale for species i on j^{th} normal line. Based on this transformation, the locations of the individual SFS profiles $n = n_p$ are aligned at $n_{i,j}^* = 0$ for all normal lines. The SFS concentration $\theta_{i,j}^s$ is then expressed as a function of the transformed variable and normalized by the magnitude of the SFS concentration $\text{mag}(\theta_{i,j}^s)$ as,

$$\hat{\theta}_{i,j}^s(n_{i,j}^*(n)) = \frac{\theta_{i,j}^s(n)}{\text{mag}(\theta_{i,j}^s)}, \quad (11)$$

where $\hat{\theta}_{i,j}^s$ refers to the normalized SFS concentration. By construction, $\hat{\theta}_{i,j}^s$ has its peak at $n_{i,j}^* = 0$ and a maximum value of unity for single banded species such that the zero-crossover locations surrounding the peak are separated by unity for single banded species. For double banded species, $\hat{\theta}_{i,j}^s$ is centered at $n_{i,j}^* = 0$ with one positive and negative peak on either side and a separation of unity between the two peaks, thus factoring out the variability from individual SFS concentration parameters.

The SFS structure is quantified statistically by ensemble averaging $\hat{\theta}_{i,j}^s(n_{i,j}^*)$ across all normal lines weighted by the flame-sheet area surrounding each normal line. This is plotted in Fig. 7 as a function of $n_{i,j}^*$ scaled by the average length scale of the SFS concentration relative to l_F . Figure 7 shows that the SFS structure of CH₂O, H₂ and HCO concentration

is single-banded in an averaged sense, and double-banded for OH, n-C₇H₁₆ and CO₂. The SFS structure of H and O concentration (not shown) is found to be double-banded in mean.

In terms of the variability in $\hat{\theta}_{i,j}^s$, all single banded species have standard deviations $\sigma(\hat{\theta}_{i,j}^s) < 0.25$ between the zero cross-over locations. CH₂O and H₂ have higher standard deviation towards the reactant side than the products side of the flame-sheet, while HCO has similar levels of standard deviation in both the pre-flame and post-flame locations. In the case of double banded species, the standard deviation is < 0.3 within the influence zone. OH has roughly equal standard deviations in both the pre-flame and post-flame locations. n-C₇H₁₆ and CO₂ have similar magnitudes of standard deviation near the flame-sheet for $|n_i^* \langle l_\theta(\theta_i^s) \rangle| \lesssim 2l_F$, but are consistently higher in the far reactants side (i.e. for $n_i^* \langle l_\theta(\theta_i^s) \rangle \lesssim -2l_F$) compared with that on the products side. These characteristics of the variability in structure of SFS concentration will be shown to be related with the parameterization variability (Section IV B 2) and interactions between flame fronts in the following section.

The results demonstrate that the SFS species concentrations are single or double banded over the flame on average with deviations that do not alter the structural form. To generalize these results for all species, the filtering procedure causes the species concentrations for species that are produced and consumed within the flame to be underestimated in the flame and overestimated outside the flame by the filtered concentrations, resulting in an single-banded structure on average. Species that exhibit a frontal variation are underestimated on one side of the flame-sheet and over-estimated on the other side of the flame-sheet resulting in a double-banded structure on average.

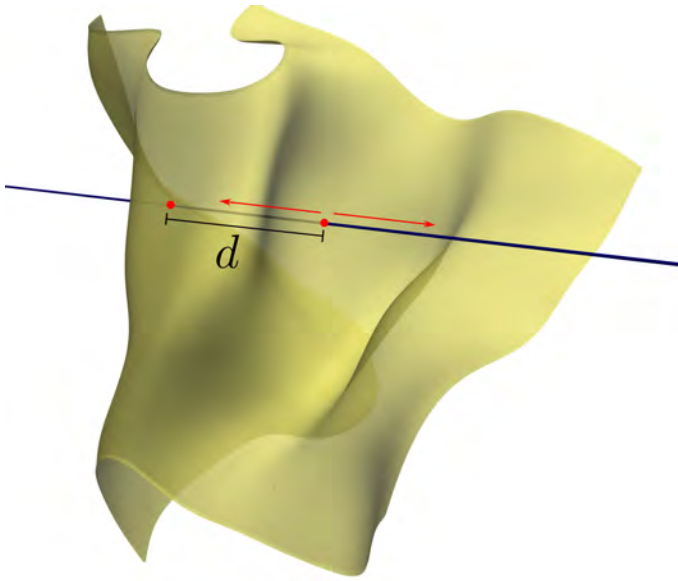


FIG. 8: Illustration of the linear normal line from a given location on the flame-sheet (yellow).

C. Relationship Between SFS Structure and Features of the Flame Surface: Interactions Between Flame Fronts and Variations in Flame Curvature

The variability in the SFS species structure could result from differences in the flame conditions from location to location, interactions between neighboring flame fronts and differences in flame curvature among other factors. We first analyze the effects of flame-flame interactions on the SFS structure variability by quantifying the integrated deviations $\varepsilon_{i,j}$ between the structure of the SFS concentrations $\hat{\theta}_{i,j}$ and the mean structure $\langle \theta_{i,j} \rangle$ from Section IV B 3 as,

$$\varepsilon_{i,j} = \sqrt{\frac{\int_{\xi_i^l}^{\xi_i^u} (\hat{\theta}_{i,j} - \langle \hat{\theta}_{i,j} \rangle)^2 dn_i^*}{\int_{\xi_i^l}^{\xi_i^u} \langle \hat{\theta}_{i,j} \rangle^2 dn_i^*}}, \quad (12)$$

where the integration limits ξ_i^l and ξ_i^u are the respective lower and upper limits of the overall influence zone as determined in Section IV A after applying the transformation given by Eq. (10).

1. Relationship with Proximity Between Neighboring Flame-sheets

To understand how $\varepsilon_{i,j}$ varies with the proximity between flame-sheets, straight lines are constructed as shown in Fig. 8 from the discrete nodes on the flame-sheet by following the direction of the initial normal to the flame-sheet i.e. with $\nabla C^r(\mathbf{y}_j) / |\nabla C^r(\mathbf{y}_j)|$ on the RHS of Eq. (7) for the j^{th} normal line. These straight lines may intersect the flame-sheet ($C^r = 0.5$) multiple times if there is a surrounding flame. As a measure of proximity between neighboring flame-sheets, distances are computed between the closest points of intersection of these straight lines to another flame-sheet on the reactant side as d_j^- and product side as d_j^+ as illustrated in Fig. 8. In the current simulation, about 40% of the flame surface area has a nearby flame-sheet within $10l_F$ on the reactant and product sides respectively.

The integrated deviation from mean, $\varepsilon_{i,j}$ is quantified separately for the left-side (i.e. $n_{i,j}^* < 0$) and right-side (i.e. $n_{i,j}^* > 0$) of $n_{i,j}^* = 0$ within the influence zone using Eq. (12) to distinguish between differences on either side, denoted as $\varepsilon_{i,j}^-$ and $\varepsilon_{i,j}^+$ respectively. For the species shown in Fig. 7, logarithm of joint PDF of $\varepsilon_{i,j}^-$ ($\varepsilon_{i,j}^+$) and d_j^- (d_j^+) weighted by the area surrounding corresponding normal lines are shown in Figures 9 and 10. From the distribution in Fig. 9(a), the integrated deviation from mean in CH₂O is likely to be smaller ($\varepsilon_{\text{CH}_2\text{O},j}^- < 0.3$) for $d_j^- \gtrsim 3.5l_F$. However, a distinct transition is observed at $d_j^- \approx 3.5l_F$ below which the most likely $\varepsilon_{\text{CH}_2\text{O},j}^-$ increases with decreasing d_j^- . The conditional mean of $\varepsilon_{\text{CH}_2\text{O},j}^-$ with d_j^- in Fig. 9(a) clearly depicts this transition. Similar trends are observed for H₂ and HCO, where the transition is observed about $d_j^- \approx 3.5l_F$ and $d_j^+ \approx 2l_F$ respectively. $\varepsilon_{i,j}^-$ is in general, smaller for HCO compared with CH₂O and H₂.

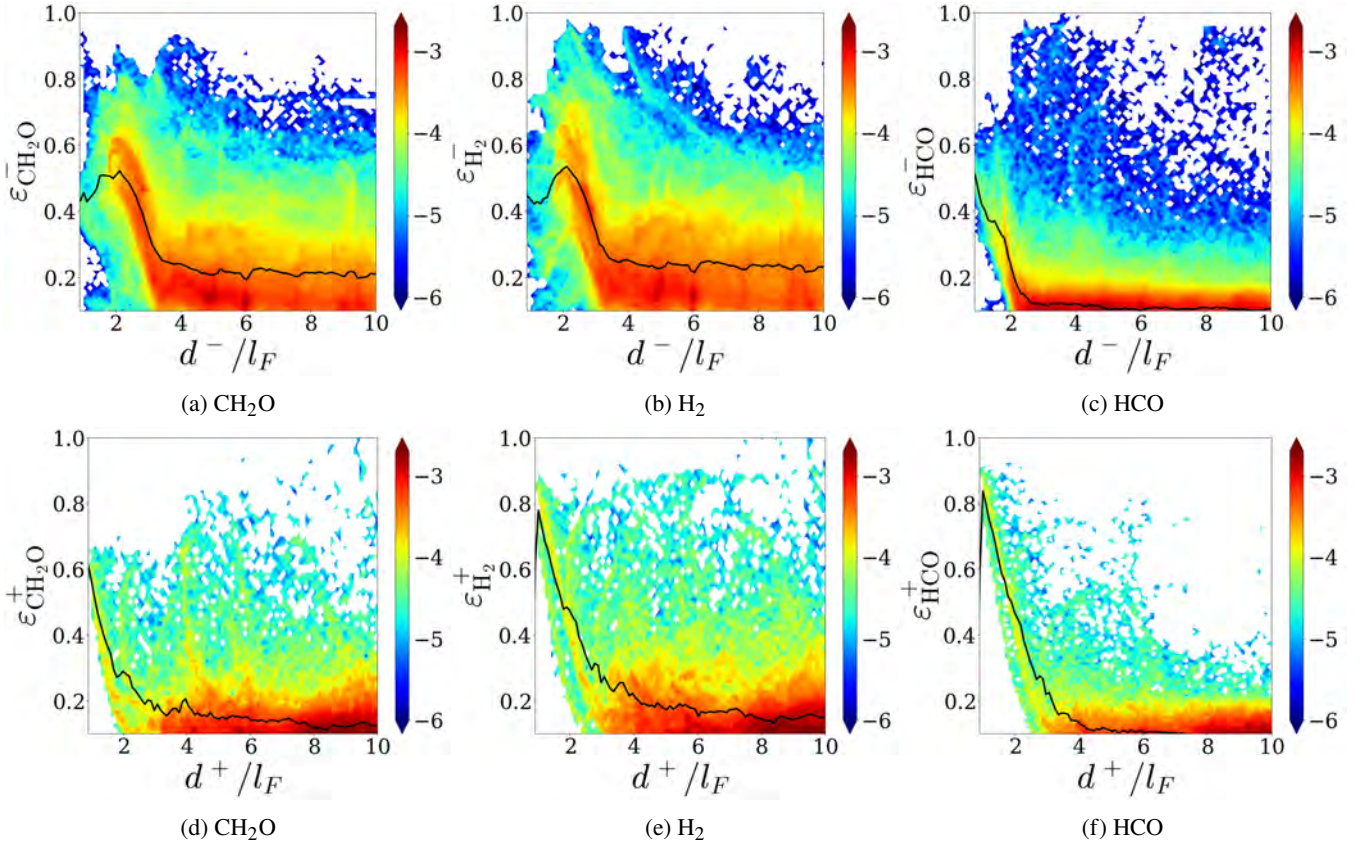


FIG. 9: Logarithm of area weighted joint PDF of $\varepsilon_{i,j}^-$ with d_j^- (top) and $\varepsilon_{i,j}^+$ with d_j^+ (bottom) for single-banded species. Each panel shows PDF variations of up to three orders of magnitude and the black lines indicates conditional mean of $\varepsilon_{i,j}^-$ ($\varepsilon_{i,j}^+$) with d_j^- (d_j^+).

For the right-side integrated deviation $\varepsilon_{i,j}^+$, the mean values are smaller compared with $\varepsilon_{i,j}^-$ for $d^+ > 4l_F$, with a distinct transition to increasing integrated deviations below $d_j^+ \approx 3l_F$ for CH_2O and H_2 and $d_j^+ \approx 4l_F$ for HCO .

Among these single-banded species, the SFS CH_2O and H_2 concentrations show higher levels of $\varepsilon_{i,j}^-$ for $n_{i,j}^* < 0$ compared with $\varepsilon_{i,j}^-$ levels for HCO . This is expected given that the subfilter-scale CH_2O and HCO concentrations have their mean locations $\langle n_p \rangle < 0$ (Fig. 6 and Table III) making them susceptible to the influence of nearby flame-sheet from the unburned reactants side. These are also the species that are distributed over a larger length scale with $\langle l_\theta \rangle \sim 1l_F$ compared with HCO ($l_\theta \approx 0.6l_F$). On the other hand, HCO has its mean location towards the product side of the flame-sheet ($\langle n_p \rangle = 0.13l_F$) in Fig. 6 with a smaller spread based on its length scale making the deviations in HCO less sensitive to the flame-sheet separation. To generalize these results, structure of single-banded species with peak locations away from the flame-sheet or with larger length scales are more susceptible to the influence of a neighboring flame-sheet i.e. higher $\varepsilon_{i,j}$ and consequently a higher standard deviation in Fig. 7.

The logarithm of the joint PDF of integrated deviations are similarly quantified for double-banded species, OH , $\text{n-C}_7\text{H}_{16}$

and CO_2 in Fig. 10. $\varepsilon_{\text{OH},j}^-$ tends to be smaller (< 0.2) for $d_j^- \gtrsim 2l_F$ compared with $\text{n-C}_7\text{H}_{16}$ and CO_2 . A distinct transition from roughly constant lower values of $\varepsilon_{\text{OH},j}^-$ in mean is observed at $d_j^- \approx 2l_F$, below which mean $\varepsilon_{\text{OH},j}^-$ increases with decreasing d_j^- . Similar trends are observed for $\text{n-C}_7\text{H}_{16}$ and CO_2 with transitions at $d_j^- \approx 3.6l_F$ and $3l_F$. On the right-side ($n_{i,j}^* > 0$), $\varepsilon_{\text{OH},j}^+$ transitions from a mean value of about 0.2 to larger values at about $d_j^+ \approx 4l_F$, while for $\text{n-C}_7\text{H}_{16}$ such transition is not present with a mean integrated deviation not exceeding 0.35 for all d_j^+ . In the case of CO_2 , mean $\varepsilon_{\text{CO}_2,j}^+$ is higher (about 0.3) for $d_j^+ > 4l_F$ and increases with decreasing flame separation on the product side for $d_j^+ < 4l_F$. $\varepsilon_{\text{CO}_2,j}^+$ is also shown to have a larger spread for $d_j^+ > 4l_F$, reaching up to a disparity level of 0.6.

These differences can be explained by considering the mean location and scale of SFS species concentrations (Table III). The SFS OH concentration is located towards the product side of the flame-sheet ($\langle n_p \rangle = 0.28l_F$) and has a smaller scale ($l_\theta = 0.76l_F$) making it more sensitive to flame-sheet separation on the product side leading to $\varepsilon_{\text{OH},j}^+ > \varepsilon_{\text{OH},j}^-$ in mean. CO_2 is located closer to the flame-sheet ($\langle n_p \rangle = 0.07l_F$), but distributed over a larger length scale ($l_\theta = 1.79l_F$) which results in similar levels of $\varepsilon_{\text{CO}_2,j}$ on each side. $\text{n-C}_7\text{H}_{16}$ is located

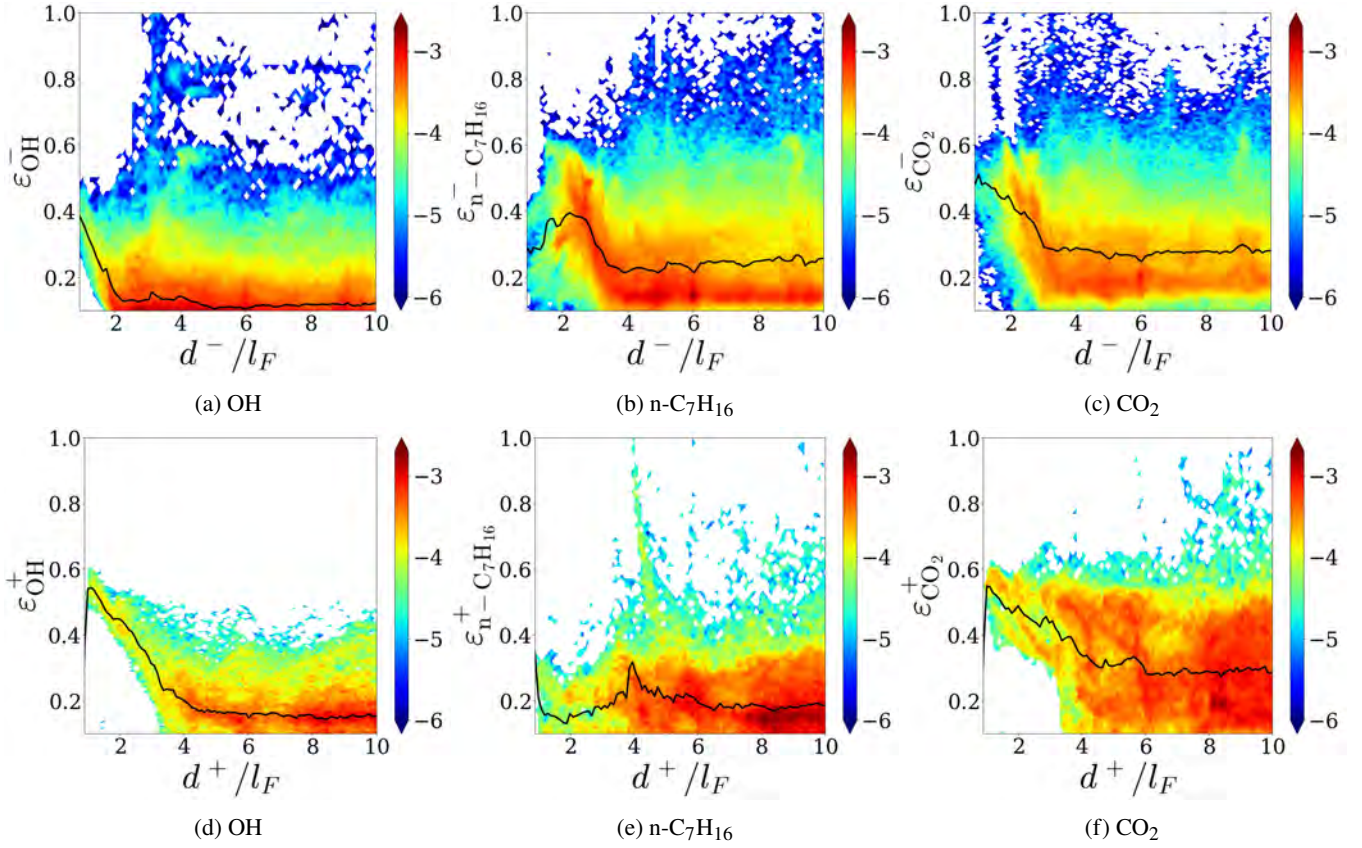


FIG. 10: Logarithm of area weighted joint PDF of $\varepsilon_{i,j}^-$ with d_j^- (top) and $\varepsilon_{i,j}^+$ with d_j^+ (bottom) for double-banded species. Each panel shows PDF variations of up to three orders of magnitude and the black lines indicates conditional mean of $\varepsilon_{i,j}^-$ ($\varepsilon_{i,j}^+$) with d_j^- (d_j^+).

on the reactants side of the flame-sheet with $\langle n_p \rangle = -0.64l_F$ and distributed over an intermediate length scale between OH and CO_2 , which makes it more sensitive to flame-sheet separation from the reactant side (d_j^-). Consequently, $\varepsilon_{\text{n-C}_7\text{H}_{16},j}^-$ is higher than $\varepsilon_{\text{n-C}_7\text{H}_{16},j}^+$ in mean with a higher sensitivity to smaller flame-sheet separation on the reactant side compared with products side. These results indicate that the structure of the species with larger length scales or placement away from flame-sheet such as n-C₇H₁₆ or CO_2 is affected to a greater extent from flame-flame interactions than species with smaller length scales such as OH. Consequently, these species with larger length scales or distant placement from flame-sheet (Fig. 6) have larger standard deviations shown previously in Fig. 7 as well.

2. Relationship with Local Flame Curvature

Although the deviations in the structure of SFS species concentration are shown to be correlated to the separation between flame-sheet up to a certain threshold separation, there is a “spread” in $\varepsilon_{i,j}$ for all separation distances as shown in Figures 9 and 10. This spread is investigated by examining the relationship of deviations in the SFS structure to the flame

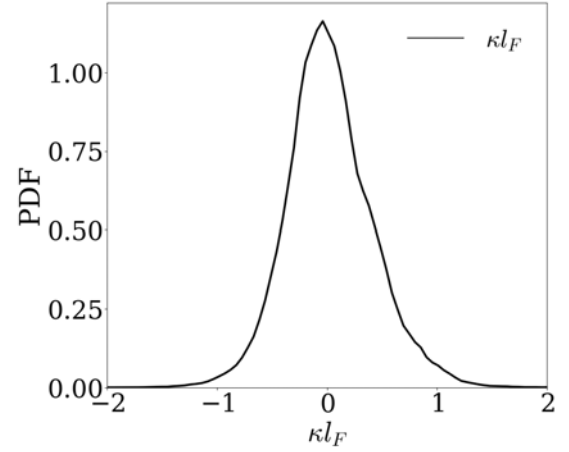


FIG. 11: PDF of the normalized algebraic curvature distribution on the flame-sheet weighted by the area surrounding each node.

curvature. The PDF distribution of the algebraic flame curvature κ normalized by the laminar flame thickness is shown in Fig. 11.

Joint PDFs are constructed between $\varepsilon_{i,j}$ and the absolute

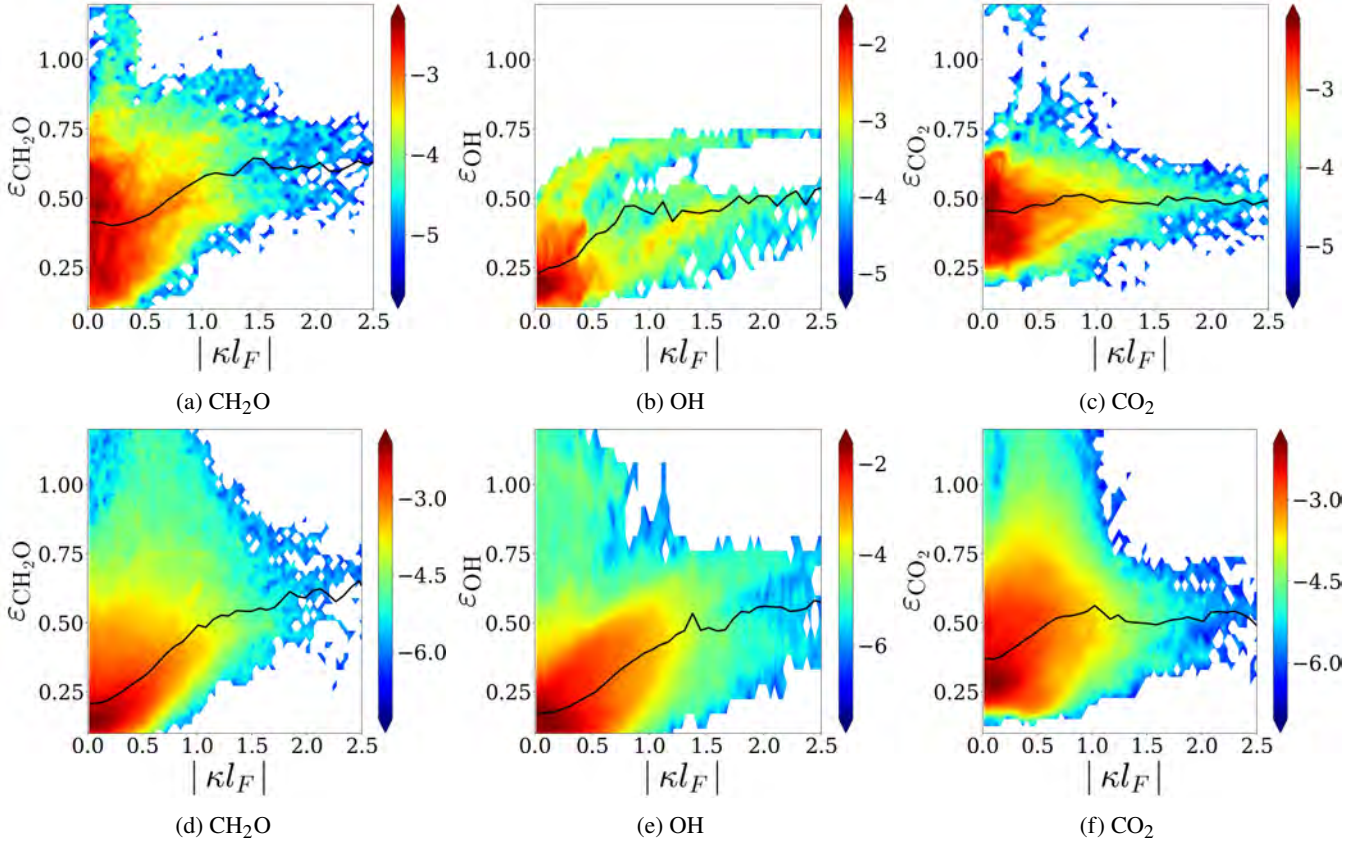


FIG. 12: Logarithm of conditional PDF distribution of $\varepsilon_{i,j}$ with algebraic curvature κ relative to l_F for one single-banded (CH_2O) and two double-banded species (OH and CO_2). Top row shows results conditioned for $d/l_F \leq 3.5, 2.0$ and 3.0 for CH_2O , OH and CO_2 respectively and bottom row shows results conditioned over remaining values of d . Black lines indicate conditional means of $\varepsilon_{i,j}$ with $|\kappa l_F|$.

curvature $|\kappa|$ for each single and double banded species to investigate the relationship between the differences in SFS structure and flame curvature. Figure 12 shows the logarithm of such PDF for select species (CH_2O , OH , and CO_2) conditioned separately over parts of flame-sheet where flame proximity effects are strong ($d \leq 3.5l_F$ for CH_2O , $d \leq 2l_F$ for OH and $d \leq 3l_F$ for CO_2). The conditional means shown by the black lines indicate an increasing trend for $\varepsilon_{i,j}$ with curvature for all species in Fig. 12. CH_2O a higher $\varepsilon_{\text{CH}_2\text{O},j} = 0.4$ in mean at lower curvatures (Fig. 12(a)) compared with disparity level of $\varepsilon_{\text{CH}_2\text{O},j} = 0.2$ (Fig. 12(d)) in regions with stronger and weaker flame proximity effects respectively. $\varepsilon_{\text{CH}_2\text{O},j}$ is consistently higher with a smaller overall change with curvature when flame proximity effects are stronger, while $\varepsilon_{\text{CH}_2\text{O},j}$ shows greater sensitivity to curvature of the flame sheet in regions with weaker flame proximity effects causing a net overall change from $\varepsilon_{i,j} \approx 0.2$ to 0.6 when going from lower to higher curvature flame regions. Similar results are obtained for the double banded species OH and CO_2 respectively. At lower curvature regions, the disparity levels in SFS OH concentration in regions with stronger and weaker flame proximity are about 0.25 and 0.1 (i.e. different by about 50%), while disparities in SFS CO_2 concentration in corresponding regions are about 0.4 and 0.3 . These disparities show an increasing trend

with curvature for OH for all d and approach $\varepsilon_{\text{OH},j} \approx 0.5$ at higher curvature levels. In comparison, the disparity in CO_2 is roughly constant with κ at $\varepsilon_{\text{CO}_2,j} \approx 0.5$ for $d \leq 3l_F$, suggesting a stronger sensitivity to flame proximity consistent with findings from Section IV C 1.

D. Discussions

Most existing modeling approaches target the unclosed terms in LES of turbulent flames, such as the SFS contributions to the scalar flux terms, $[\nabla \cdot (\rho \theta_i \mathbf{u}) - \nabla \cdot ((\rho \theta_i)^r \mathbf{u}^r)]^r$, using, for instance, eddy-diffusivity-based models²⁴, and chemical reaction rates, $w^r(T, \theta_i)$, using, for instance, flamelet-based models³⁶. Our work explores the possibility for a new approach in which the SFS temperature field (T^s) and SFS species concentrations (θ_i^s) are approximated with analytical expressions to reconstruct the unclosed terms mentioned above. This strategy could be advantageous since, once developed, the models for SFS quantities (T^s, θ_i^s) can be applied to both unclosed scalar flux terms and chemical reaction rates. Moreover, directly modeling these SFS quantities would, in principle, allow the LES solution to approach the DNS solution progressively with grid refinement. Results reported in this

study on the structure of SFS scalar fields could be used with a database to parameterize mathematical forms for SFS quantities in terms of placement, scale and magnitude relative to the predicted resolved-scale flame location and topology. These analytical functions would be designed based on the knowledge of the corresponding RS fields to ensure the physical realizability of the full fields (e.g., $T^s \geq -T^r$ and $\theta_i^s \geq -\theta_i^r$).

Our analysis is carried out for a single filter width, $\Delta = 2l_F$, which was determined so that momentum and energy have at least 85% of the total variance in the resolved scales. This filter width is generally practical and affordable in LES of turbulent premixed flames, whereas smaller filter widths (e.g., $\Delta = l_F$) are generally not achievable in practice². We believe that the basic single- and double-banded structure observed for SFS scalar profiles along flame normal lines is independent of filter width. As described in Section IV B, full species concentrations that peak within the flame are under-estimated by the resolved-scale (filtered) concentrations around the peak and over-estimated away from the peak, leading to the characteristic single-banded structure in SFS species concentration. Full species concentrations that display a frontal behavior across the flame, on the other hand, are under-estimated and over-estimated by the resolved-scale (filtered) concentrations on each side of the front, which results in the characteristic double-banded structure in SFS species concentrations. A different filter width is expected to affect the length scale and magnitude of SFS quantities, but not the qualitative SFS behaviors described above. However the specific upper and lower limits of the influence zones may depend on filter width. With increasing filter width, the resolved-scale temperature and species concentrations are expected to spread over larger length scales and the corresponding subfilter-scale temperature and species concentrations are expected to have larger magnitudes over broader regions surrounding the flame. As a result, chemical reaction rates estimated from resolved-scale quantities, $w_{i,j}(T^r, \theta^r)$, are expected to have larger deviations from the actual reaction rates, $w_{i,j}(T, \theta)$, over larger regions, which leads to larger zones of influence with increasing filter width. Conversely, the zones of influence are expected to be smaller with decreasing filter width.

V. CONCLUSIONS

An *a priori* analysis of subfilter-scale(SFS) species structure important to the estimation of chemical reaction rate nonlinearity in large-eddy simulation (LES) is performed using direct numerical simulation (DNS) of a turbulent premixed flame. Curvilinear normal lines originating from the flame-sheet are constructed to identify “zones of influence” within which the incorporation of the SFS content has an impact to the estimation of the chemical reaction rates in LES, and to analyze the structure of SFS species concentrations. Using this framework, the reaction specific zones of influence are found to be localized within $3.3l_F$ from the flame front on average. Within these zones of influence, the SFS species concentrations are shown to conform to two structural groupings referred to as “single-banded” and “double-banded” structure.

Species structure located towards the unburned reactants side of the flame is found to be influenced more strongly from interactions with neighboring flame-sheets on the reactant side. These species show higher standard deviations from the mean SFS structure on the unburned reactant side of the flame. Species structure characterized by smaller length scales or with placement closer to the flame-sheet are shown to have similar levels of standard deviation on both sides of the flame-sheet. The structure of these species is shown to be equally susceptible to modification from interactions with neighboring flame-sheet from the pre-flame and post-flame sides. The deviations in local SFS structure from the mean SFS structure are shown to increase with flame curvature for all species.

These results are shown to rely upon the specific characteristics pertaining to the behavior (e.g. frontal variation vs. species being produced and consumed within the flame), placement and scales for several different species types, including initial reactants and final combustion products as well as intermediate species and free radicals, participating in a diverse set of reactions from the chemical model. We expect these results to be generalizable to other species and reactions showing similar characteristics within these types. In particular, species that are produced and consumed within the flame similar to those considered in this study are expected to show single-banded structure, while those that display a frontal behavior are expected to show double-banded structure. Species concentrations that peak away from the flame front, such as concentrations associated with initial reactants or final combustion products show greater standard deviations in their mean SFS structure due to flame-flame interactions. Species with larger chemical time-scales that are spread over larger length scales are affected by interactions from a nearby flame front to a greater extent and display higher standard deviation from their mean SFS structure. On the other hand, radical species that are formed closer to the flame front and with shorter chemical time-scales are likely less affected from interactions with neighboring flame-front and display smaller standard deviations from their mean SFS structure.

As the conclusions derived from this study depend upon the characteristics of sub-filter scale structure and variations resulting from local interactions over a range of flame curvatures and flame-flame interactions, they may be expected to display a certain degree of universality within the tested conditions in the thin flames regime. At other flame conditions, such as those with significantly higher Karlovitz numbers, we expect the higher curvature regions to become more prominent. Under such conditions, stronger deviations in structure of SFS species concentrations may be expected from the mean SFS structure. These results may be expected to be generalizable to other filter widths to show varying degrees of deviations in the distribution of SFS structure. At smaller filter widths, the differences between the LES-filtered quantities and those computed in DNS would be confined to smaller scales. These differences would diminish progressively as the filter width approaches DNS resolution and the corresponding zones of influence and scales of SFS structure are expected to become smaller with decreasing filter widths. Conversely, larger zones

of influence may be expected at larger filter-widths as the SFS content is distributed over larger scales. This may make the SFS structure more susceptible to impacts from flame-flame interactions at larger filter-widths.

SUPPLEMENTARY MATERIAL

See supplementary material for a complete list of the resolved-scale variance fractions of species concentrations.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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