TURBULENT CHEMICAL AND THERMONUCLEAR FLAMES:
INTRINSIC INSTABILITY AND ANISOTROPIC TURBULENCE
AMPLIFICATION

Alexei Y. Poludnenko, Brian D. Taylor
Naval Research Laboratory
Washington, D.C. 20375 U.S.A.
alexei.poludnenko@nrl.navy.mil

ABSTRACT

In our previous study (Poludnenko, 2015), we presented the analysis of the intrinsic stability of high-speed turbulent reacting flows. A systematic survey of a wide range of turbulent intensities and system sizes showed that turbulent flames in the regimes considered are intrinsically unstable even in the absence of the surrounding combustor walls or obstacles, which can support the thermoacoustic feedback. In particular, three effects were observed. 1) The turbulent flame speed, $S_T$, can develop pulsations with the observed peak-to-peak amplitude $S_T^{\max}/S_T^{\min} > 10$. 2) Unstable burning results in the periodic pressure build-up and the formation of pressure waves or shocks, when $S_T$ approaches or exceeds the speed of a Chapman-Jouguet deflagration. 3) Coupling of pressure gradients formed during pulsations with density gradients across the flame leads to the anisotropic amplification of turbulence inside the flame volume and flame acceleration.

In this work we extend prior analysis, which relied on a simplified single-step reaction model, by demonstrating existence of the pulsating flame instability in two realistic reactive systems: chemical flames in atmospheric H₂-air mixtures and thermonuclear flames in degenerate, relativistic plasmas found in stellar interiors. Finally, we also consider the dependence of the instability on the system size by performing a direct numerical simulation containing 32 billion cells in a domain twice larger than considered by Poludnenko (2015). No significant change in the instability dynamics is observed, though further analysis of this question for different Karlovitz and Damköhler numbers is required.

INTRODUCTION

Hydrodynamic flows containing exothermic reaction fronts, or flames, exhibit a rich variety of unstable phenomena. Detailed understanding of such instabilities is of significant practical importance since, ultimately, they critically control the dynamics of burning in the host system. Of particular interest here are intrinsic flame instabilities, which are independent of the external factors such as the overall geometry of the combustor, since such instabilities are generally much more difficult to control in a practical setting. In this context, historically, the primary focus of both theoretical and experimental studies has been on various hydrodynamic (e.g., Landau-Darrieus), thermodiffusive (e.g., cellular), and body-force (e.g., Rayleigh-Taylor) instabilities of premixed laminar flames (Williams, 1985).

Realistic combustion systems, such as an aircraft jet engine, however, generally rely on turbulent flames for their operation. The question of turbulent flame stability has been primarily considered in the context of burning in confined environments with walls or obstacles. In such situations, thermoacoustic instabilities result from the resonant coupling between the exothermic process and the acoustic field, which it generates in the interior of a combustor (Poinset & Veynante, 2005; Kadowaki & Hasegawa, 2005; Candel et al., 2013). A critical aspect here is that such instabilities, which are intimately tied to the host system, can potentially be efficiently suppressed, or even completely eliminated, e.g., by changing the combustor geometry.

At the same time, prior direct numerical simulations (DNS) of turbulent premixed flames suggest that such flames can exhibit significant variations in their key dynamical characteristics, in particular flame structure and burning speed, even in the absence of the surrounding combustor walls and under the most idealized circumstances of a statistically steady, homogeneous, isotropic upstream turbulence. For instance, DNS by Nishiki et al. (2002) and Bell et al. (2006) modeled flame interaction in an unconfined domain with a relatively low intensity turbulence with Damköhler number $Da \approx 17 – 18$ in Nishiki et al. (2002) and $Da \approx 1.5$ in Bell et al. (2006), and with Karlovitz number $Ka = 1$ in both cases. The maximum peak-to-peak amplitude of the turbulent burning velocity $S_T^{\max}/S_T^{\min} \lesssim 2.0$ was observed. In contrast, in our earlier work (Poludnenko & Oran, 2010, 2011), we considered a much higher intensity turbulence with the integral velocity $U_I = 18.5 S_T$ and scale $l = 1.9 \delta_T$, where $S_T$ and $\delta_T$ are, respectively, the speed and the thermal width of a laminar flame (cf. case 6 in Fig. 1). Even though $U_I$ in the upstream flow varied only by a few percent over the course of the calculation, resulting turbulent flame speed exhibited significant variability with $S_T^{\max}/S_T^{\min} \approx 3$.

Recently, we carried out a systematic investigation of the intrinsic stability of premixed turbulent flames in the thin reaction zones regime (Poludnenko (2015), hereafter P15). In particular, that study surveyed a wide range of turbulent conditions, which are marked as cases 14 – 18 in the classical combustion regime diagram (Fig. 1). The key result of that study was the demonstration that turbulent flames can indeed exhibit significant variability of $S_T$. This is illustrated for cases S18 and S16 (P15) in Figs. 2a and 3a (also see Table 1). For instance in S16, $S_T$ was observed to vary by more than an order of magnitude in the course of the simulation. The primary source of...
such instability is the resonant state between the continuous creation of the flame surface by turbulence and its intermittent rapid destruction by flame collisions. This process is illustrated in Fig. 4, which shows the change in the flame structure in the course of a typical pulsation. As shown in P15, with increase in turbulent intensity \( U_l > S_L \), flame surface consumption through flame self-propagation becomes progressively less important. As a result, in the absence of flame collisions (Fig. 4a), the flame surface grows exponentially under the action of turbulence, as it would for a passively advected interface. Such flame folding makes the flame surface more convolved and greatly increases the overall burning rate. At the same time, it also results in the eventual formation of extended regions of flame collision (Fig. 4b), which rapidly consume the flame surface. This restores a more planar flame configuration seen in Fig. 4a decreasing \( S_T \) and, thus, setting up the next pulsation cycle.

It was observed in P15 that the magnitude of this instability depends non-monotonically on turbulent intensity reaching maximum strength at some intermediate values of \( U_l / S_L \). In particular, when \( U_l < S_L \), flame self-propagation is important and it attenuates the exponential growth of the flame surface. In contrast, at high turbulent intensities \( U_l > S_L \), the flame is extremely tightly packed. This leads to frequent flame collisions, which interrupt the periods of exponential flame-surface growth and thus prevent the development of large-amplitude pulsations.

The second finding reported in P15 was the fact that such flame pulsations can result in a periodic formation of strong pressure pulses and even shocks. In particular, formation of shocks was observed with \( M_a > 1.3 \) in the course of flame interaction with \( M_{at} = 0.02 \) upstream turbulence. This is illustrated in Fig. 2a for case S18, which shows the formation of overpressures as high as 40%.

Formation of such pressure pulses occurs when \( S_T \) becomes a significant fraction of, or exceeds, the speed of a Chapman-Jouguet deflagration, \( S_{CJ} \) (Williams, 1985). We showed in an earlier work (Poludnenko et al., 2011) that \( S_{CJ} \) serves as a threshold, at which the amount of energy generated inside the flame volume on its sound-crossing time becomes comparable to the internal energy of the fluid. This, in turn, results in the build-up of pressure, which can ultimately produce a catastrophic runaway process ending in a spontaneous deflagration-to-detonation transition (DDT). In fact, the ratio \( CJ = S_T / S_{CJ} \) is effectively a reactive-flow counterpart of the Mach number. It serves as a figure of merit characterizing the importance of compressibility effects in a reaction wave, i.e., a flame, rather than a hydrodynamic wave as in the case of Mach number.

It is important to emphasize that, as shown in P15 and as will be demonstrated below, the onset of the pulsating instability of \( S_T \) does not require the formation of large overpressures. In fact, the latter are the consequence, rather than the cause, of the former. In those cases where the flame does produce overpressures, they can result in a significant amplification of turbulence inside the flame brush. This process can, in turn, alter the overall flame dynamics.

The mechanism of turbulence amplification is analo-
Figure 3. Comparison of the effect of a single-step and detailed H$_2$ kinetics on the pulsating instability. Shown are the key dynamical characteristics of the flame in simulations S16 (Poludnenko, 2015) and S16H$_2$ (see caption in Fig. 2 for the description). Note that the horizontal and left vertical axes in both panels have the same scale to facilitate comparison between the two cases.

Figure 4. Change in the flame structure in the course of a typical flame pulsation in the calculation using single-step Arrhenius kinetics (case 16s described in Poludnenko (2015)). Shown is the flame volume bounded by the two isosurfaces of the fuel mass fraction $Y = 0.05$ (red) and $Y = 0.95$ (blue) at $t = 16.72\tau_{ed}$ (panel a) and $t = 19.13\tau_{ed}$ (panel b). These instances approximately correspond to the times of minimum and maximum flame burning velocity (reproduced from Poludnenko (2015)).

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Table 1. Summary of calculations performed

<table>
<thead>
<tr>
<th>Grid</th>
<th>( \frac{\tau_S}{\tau_{ed}} )</th>
<th>( Da )</th>
<th>( CI_L )</th>
<th>( \frac{l}{\delta_L} )</th>
<th>( \frac{U_l}{S_L} )</th>
<th>( \frac{\Sigma_f}{S_L} )</th>
<th>( \frac{\Sigma_{min}}{S_L} )</th>
<th>( \frac{P_{max}}{S_{min}} )</th>
<th>( \tau_{ed} )</th>
<th>( \frac{\delta U_1}{S_L} )</th>
<th>( \frac{\delta U_2}{S_L} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>S16</td>
<td>256^2 \times 8,192</td>
<td>26.76</td>
<td>0.60</td>
<td>0.06</td>
<td>3.73</td>
<td>6.23</td>
<td>7.89</td>
<td>10.42</td>
<td>1.43</td>
<td>3.33</td>
<td>6.70</td>
</tr>
<tr>
<td>S16H2</td>
<td>256^2 \times 8,192</td>
<td>21.89</td>
<td>0.60</td>
<td>0.04</td>
<td>3.71</td>
<td>6.22</td>
<td>12.05</td>
<td>7.19</td>
<td>1.39</td>
<td>2.78</td>
<td>6.37</td>
</tr>
<tr>
<td>S18</td>
<td>512^2 \times 16,384</td>
<td>10.05</td>
<td>5.94</td>
<td>0.06</td>
<td>14.92</td>
<td>2.49</td>
<td>9.01</td>
<td>4.34</td>
<td>1.31</td>
<td>0.91</td>
<td>8.52</td>
</tr>
<tr>
<td>S19v</td>
<td>1.024^2 \times 32,768</td>
<td>2.55</td>
<td>4.67</td>
<td>0.06</td>
<td>29.35</td>
<td>3.14</td>
<td>11.78</td>
<td>2.78</td>
<td>1.26</td>
<td>1.03</td>
<td>10.90</td>
</tr>
<tr>
<td>S20k</td>
<td>512^2 \times 8,192</td>
<td>10.41</td>
<td>7.20</td>
<td>0.008</td>
<td>19.69</td>
<td>2.74</td>
<td>4.53</td>
<td>2.71</td>
<td>1.01</td>
<td>2.07</td>
<td>2.04</td>
</tr>
</tbody>
</table>

See text for the definitions of various quantities. Note that for calculations S16 and S18, time-averaged values given here correspond to a somewhat larger time interval than in Poludnenko (2015). In particular, the averaging time interval here starts at \( 1\tau_{ed} \), while in Poludnenko (2015) it started at 5.5\( \tau_{ed} \).

1 MODEL AND METHOD

Turbulence-flame interactions are modeled using compressible reactive-flow equations solved using a fully unsplit corner transport upwind scheme with the PPM spatial reconstruction and the HLLC Riemann solver (Gardiner & Stone, 2008) implemented in the code Athena-RFX (Stone et al., 2008; Poludnenko & Oran, 2010). Turbulence is driven using a spectral method, which introduces in the flow divergence-free velocity fluctuations with a prescribed energy injection spectrum and rate (Poludnenko & Oran, 2010). This approach ensures that the turbulent integral velocity, \( U_l \), and scale, \( l \), in the upstream flow are nearly constant both in space and with a standard deviation of \( \lesssim 2\% \) and \( \lesssim 5\% \), respectively. Detailed analysis of the resulting turbulence, both reacting and non-reacting, including comparison with prior experimental and DNS results, was presented in Hamlington et al. (2011, 2012).

Here we consider three reaction kinetics models. The first model is the one used in our previous studies (Gamezo et al., 2008; Poludnenko, 2015). It uses a single-step, first-order Arrhenius kinetics. Equation of state is that of an ideal gas. A simplified reaction-diffusion model represents stoichiometric \( H_2 \)-air mixture under \( Le = 1 \) conditions with model parameters calibrated to reproduce correct laminar flame and detonation properties.

Next we consider a detailed multi-step reaction model for hydrogen combustion based on the 2014 San Diego mechanism (Sánchez & Williams, 2014). This mechanism includes 21 reactions involving 8 reacting species \( H, H_2, O, O_2, OH, H_2O, HO_2, \) and the inert \( N_2 \). Thermodynamic functions of pure chemical species are computed from the NASA seven-coefficient format (McBride et al., 1993). Pure species shear viscosity coefficients and binary diffusion coefficients are evaluated from the standard kinetic theory expressions (Hirschfelder et al., 1954). Thermal conduction coefficients of pure species are evaluated using expressions given by Warnatz (1982). Mixture-averaged conduction and shear viscosity coefficients are computed from averaging formulas of order 1/4 and order 6, respectively, as recommended by Ern & Giovangigli (1994). Mixture-averaged diffusion coefficients for each species are computed as in the TRANSPORT library (Kee et al., 1986). In the interest of run-time performance, specialized source code for the chemical reaction source terms is generated from a CHEMKIN (Kee et al., 1996) input file in a preprocessing step and subsequently compiled into Athena-RFX. The preprocessor generates code that evaluates the rates of change of species mass fractions and temperature due to chemical reactions and their analytical Jacobian generally following the formulation of Perini et al. (2012). Also generated by the preprocessor is a function that tabulates forward and reverse reaction rate constants and their temperature derivatives at start-up, so that they can be evaluated by interpolation. At run time, Athena-RFX reads CHEMKIN and TRANSPORT input files to obtain thermodynamic and transport property data. Polynomials for the mass-specific internal energy and constant-volume heat capacity are directly evaluated to initialize the solution and to solve for temperature given the conservative solution. Pure species transport properties are tabulated at start-up and subsequently evaluated by interpolation within Athena-RFX.

Finally, the third kinetics model that we consider represents thermonuclear burning in relativistic, degenerate plasmas, which are representative of conditions in stellar interiors during supernova explosions (Hillebrandt & Niemeyer, 2000). In particular, Athena-RFX implements an \( \alpha \)-chain network (Khokhlov et al., 1997), which includes the triple-\( \alpha \), \( \alpha \)-capture, and heavy-ion reactions for the following 13 isotopes: \( ^4He, ^{12}C, ^{16}O, ^{20}Ne, ^{24}Mg, ^{28}Si, ^{32}S, ^{36}Ar, ^{40}Ca, ^{44}Ti, ^{48}Cr, ^{52}Fe, \) and \( ^{56}Ni \). Nuclear reaction rates are based on the tabulation by Caughlan & Fowler (1988) with screening corrections. Equation of state includes contributions from ideal ions, degenerate electrons, radiation, and electron-positron pairs (Timmes & Swesty, 2000). Thermal conduction includes both electron and photon components with the appropriate treatment of the degeneracy effects (Timmes, 2000). In the interest of computational efficiency, both the equation of state and thermal conduction are tabulated and use bi-quadratic run-time interpolation. Since in thermonuclear flames \( Le \rightarrow \infty \) and \( Pr \rightarrow 0 \), diffusion and viscosity were not included in the physical model.

Treatment of general equations of state in Athena-RFX is implemented using the energy relaxation method of Coquel & Perthame (1998). The stiff system of equations both for chemical and thermonuclear kinetics is integrated using a non-iterative, single step, semi-implicit ODE integra-
tor YASS (Khokhlov et al., 2012). It does not employ any approximations to the Jacobian matrix, conserves species mass fractions and total energy explicitly, and provides an excellent balance between accuracy and efficiency, which is critical for large-scale DNS.

All calculations are performed in a rectangular domain with a uniform Cartesian grid. Calculations S16, S18, and S19v use single-step kinetics, while calculation S16H2 uses detailed H2 kinetics (Fig. 1). Note that in contrast to cases S16 and S18, both cases S16H2 and S19v include temperature-dependent viscosity. Case S20 uses thermal nuclear kinetics. The flow is initialized with a uniform temperature \( T_0 \) and pressure \( P_0 \). In particular, in cases S16, S19, and S19v \( T_0 = 293 \) K, and in case S16H2 \( T_0 = 300 \) K. In all of these calculations, \( P_0 = 1.01 \times 10^6 \) erg/cm\(^3\) and initial mixture properties correspond to stoichiometric H2-air.

In the thermonuclear case, \( T_0 = 10^8 \) K and \( P_0 = 2.2 \times 10^{25} \) erg/cm\(^3\), and the fuel density is \( 10^8 \) g/cm\(^3\). The initial mixture is pure \( ^{12} \)C.

Kinetic energy is injected at the scale of the domain width, \( L \), with a constant rate for the duration of a simulation. Resulting turbulent flow in the upstream cold fuel is homogeneous and isotropic with an equilibrium Kolmogorov energy spectrum \( \sim k^{-5/3} \) in the inertial range extending to scales \( \lesssim \delta_k \). Boundary conditions are periodic in the \( y \) and \( z \) directions and zero-order extrapolations in the direction of flame propagation. The absence of unphysical effects due to the boundary conditions in our numerical approach, in particular, pressure wave reflections from the upstream/downstream boundaries, is demonstrated in P15.

In calculations S16 and S16H2, grid resolution is \( \Delta x = \delta_k / 16 \), while in cases S18 and S19v, \( \Delta x = \delta_k / 8 \). With this resolution choice, in S16H2 turbulence is somewhat under-resolved in the upstream cold flow with the Kolmogorov scale \( \eta_k \approx 0.4 \Delta x \) and is well resolved in product with \( \eta_k \approx 4.4 \Delta x \). In case S19v, turbulence is well resolved throughout the domain with \( \eta_k \approx 2.2 \Delta x \) and \( \eta_k \approx 28 \Delta x \) in the thermonuclear case S20t. \( \Delta x = \delta_k / 6 \), where \( \delta_k \) is the width of the \( ^{12} \)C-burning zone. Such resolution captures the speed of a laminar flame with an accuracy of a few percent.

In all cases, nonreacting turbulence was allowed to evolve in the domain over time periods between \( \approx 2.5 \) and 5 eddy turnover times on the integral scale, \( \varepsilon_p \), before the flame was initialized. Subsequent one eddy turnover time after ignition was excluded from analysis to allow the flame to become fully developed. Table 1 shows the time interval \( t_f \) used for analysis in each case.

### 2 RESULTS AND DISCUSSION

Figure 2b shows development of the pulsating instability in case S19v. This calculation has twice larger domain size, and thus integral scale, than case S18 described in P15. Karlovitz number is the same in both cases \( \approx 52 \) (see discussion of the definition of \( K_a \) in P15). An extremely large grid with 32 billion elements, which was used in case S19v, resulted in a significant computational cost of this DNS. In particular, this calculation required \( \approx 22 \) million CPU hours on 65,536 cores on Garnet at the ERDC computing center. As a result, flow evolution in this case was followed over a shorter period of time compared to case S18.

Overall, both cases S18 and S19v show similar dynamics despite the fact that S19v has twice larger domain and also includes temperature-dependent viscosity with well resolved Kolmogorov scale throughout the domain. It can be seen that case S19v also develops pronounced pulsations. Peak burning velocities in both cases are comparable and they somewhat exceed \( S_{CJ} \). This also results in comparable peak overpressures of \( \approx 40\% \). Turbulence inside the flame brush in case S19v also undergoes strong anisotropic amplification with the time-averaged streamwise component \( \langle \overline{\Delta U_z} \rangle \), exceeding the average transverse components \( \langle \overline{\Delta U_x} \rangle \) by a factor \( 2.5 \) (Table 1). In both cases, transverse velocity components generally remain between \( U_l \) and \( U_{rms} \) in the upstream flow. Amplified turbulence inside the flame brush results in somewhat larger values of \( \overline{\Delta U_z} \) in case S19v compared to S18, namely \( 11.7 \) vs \( 9.0 \) \( \% \). This, however, may not be statistically significant resulting from a shorter time-averaging interval.

Several conclusions emerge from comparison of cases S18 and S19v. First, case S19v confirms that the presence of temperature-dependent viscosity with a fully resolved Kolmogorov scale does not have a pronounced effect on the instability. This observation was previously made in P15 based on a DNS with a 4 times smaller domain containing a somewhat under-resolved upstream turbulence. Second, doubling of the domain size compared to case S18 does not appear to increase the amplitude of either \( S_f \) or \( P \). Note, however, that while \( K_a \) is the same in both calculations, \( D_a \) is somewhat smaller in case S19v. This results in a stronger role played by flame self-propagation, which may attenuate the instability. We are currently performing a calculation similar to S19v, but which has the same \( D_a \) as S18. This calculation along with S19v will show the relative impact of large-scale (or \( D_a \)) vs. small-scale (or \( K_a \)) turbulence on the development of the pulsating instability. Results of this additional DNS will be presented in a separate paper.

Comparison of cases S16 and S16H2 (Fig. 3) shows that flame pulsations are present even in the case of detailed multi-step chemistry with a realistic equation of state and transport processes. The overall amplitude of pulsations of \( S_f \) and \( P \) is somewhat smaller in S16H2, while the time-averaged \( \overline{\Delta U_z} \) is larger (also see Table 1). In particular, note that in S16, \( \overline{\Delta U_z} \) is closer both to the upstream \( U_l \) (lower boundary of the shaded gray region) and turbulent velocity fluctuations inside the flame brush (green lines in Fig. 3a). In contrast, turbulence in case S16H2, while \( \langle \overline{\Delta U_z} \rangle \) are also close to \( U_l \), \( \overline{\Delta U_z} \) is larger and is practically equal to \( U_{rms} \) in the upstream flow. The cause of such higher burning velocity in the case of detailed chemistry requires further investigation.

Finally, Fig. 5 shows the evolution of \( S_f \), \( P \), and \( \langle \overline{\Delta U_z} \rangle \) in the case of a turbulent thermonuclear flame (case S20t). Turbulent conditions in this calculation, namely
l/\delta_x, U_\delta /S_\delta, and Da are very similar to S18. While the pronounced pulsations of the burning velocity are present in S20t, their amplitude is smaller than in case S18 (Table 1). This agrees with the observation previously made in P15 that the strength of the instability diminishes with the density ratio across the flame, \( \alpha \). In particular, this effect was shown in P15 for an artificial single-step reaction-diffusion model with \( \alpha \) decreased by a factor of 2 compared to case S18. Thermonuclear flames are characterized by values of \( \alpha \lesssim 2 \) due to the degeneracy of the electron gas (Timmes & Woosley, 1992). For instance, in case S20t, \( \alpha = 1.87 \). Thus, calculation S20t corroborates the dependence of the instability on \( \alpha \) for a realistic reactive system. It is not clear, however, whether increase in the system size can partially offset the effect of \( \alpha \) and increase the amplitude of pulsations. Note also that here, similar to cases S16H2, S7 is somewhat larger than both the upstream \( U_\delta \) and \( \langle \delta U_\delta \rangle \), though \( S_\delta \) mostly remains between \( U_\delta \) and \( U_\text{rms} \) in the upstream flow. Finally, due to the extremely low value of \( C_{f_\delta} = 0.008 \), turbulent thermonuclear flame produced extremely weak pressure fluctuations on the order of \(< 1\% \).

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