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### Influence of small-scale turbulence on internal flamelet structure

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Direct numerical simulation data obtained from a highly turbulent (Kolmogorov length scale is less than a laminar flame thickness by a factor of about 20) lean hydrogen-air complex chemistry flame are processed, with the focus of the study being placed on flame and flow characteristics conditioned to instantaneous local values  $c_F(\mathbf{x},t)$  of the fuel-based combustion progress variable. By analyzing such conditioned quantities, the following two trends are documented. On the one hand, magnitudes of fluctuations of various local flame characteristics decrease with increasing the combustion progress variable, thus, implying that the influence of small-scale (when compared to the laminar flame thickness) turbulence on internal flamelet structure is reduced as the flow advance from unburned reactants to combustion products. On the other hand, neither local turbulence characteristics (conditioned rms velocities, total strain, and enstrophy) nor local characteristics of flame-turbulence interaction (flame strain rate) decrease substantially from the reactant side to the product side. To reconcile these two apparently inconsistent trends, the former is hypothesized to be caused by the following purely kinematic mechanism: residence time of turbulence within a large part of a local flamelet is significantly shortened due to combustion-induced acceleration of the local flow in the direction normal to the flamelet. This residence-time reduction with increasing  $c_F$  is especially strong in the preheat zone ( $c_F < 0.3$ ) and the residence time is very short for  $0.3 < c_F < 0.8$ . Therefore, small-scale turbulence penetrating the latter zone is unable to significantly perturb its local structure. Finally, numerical results that indirectly support this hypothesis are discussed.

Keywords: Premixed turbulent combustion; Thermal expansion; Turbulence; Flame broadening; DNS; Hydrogen

### 25 I. INTRODUCTION

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Since the pioneering work by Damköhler<sup>1</sup> and Shelkin,<sup>2</sup> substantial progress was reached in turbulent 26 combustion modeling, e.g., see papers published recently in this journal.<sup>3-12</sup> Nevertheless, the 27 28 combustion community has been striving to uncover governing physical mechanisms of the influence of turbulence on a premixed flame under different conditions. This goal was often pursued by 29 introducing combustion regime diagrams<sup>13-15</sup> where different physical scenarios were hypothesized for 30 different non-dimensional turbulent flame characteristics such as rms velocity  $u'/S_L$ , an integral length 31 scale  $L/\delta_L$ , Damköhler number  $Da = \tau_t/\tau_f$ , or Karlovitz number  $Ka = \tau_f/\tau_K$  or  $(\delta_L/\eta_K)^2$ . Here,  $S_L$ , 32  $\delta_L$ , and  $\tau_f = \delta_L/S_L$  designate laminar flame speed, thickness, and time scale, respectively;  $\tau_t = L/u'$ 33 and  $\tau_K$  are turbulence and Kolmogorov<sup>16,17</sup> time scales, respectively; and  $\eta_K$  is Kolmogorov length 34 scale. For large-scale and weak turbulence associated commonly with  $L/\delta_L \gg 1$ ,  $u'/S_L = 0(1)$ ,  $Da \gg$ 35 1, and Ka < 1, there is consensus that the influence of turbulence on a premixed flame consists 36 primarily in wrinkling flame surface, thus, increasing its area and bulk burning rate.<sup>18-20</sup> For intense 37 turbulence characterized by a small Da, a large  $(\delta_L/\eta_K)^2$ , and  $u'/S_L \gg 1$ , there is no consensus and 38

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different scenarios are still discussed. Under such conditions, large-scale turbulent structures, i.e., 39 40 structures whose length scale is substantially larger than the thickness  $\delta_L$ , are still considered to increase 41 flame surface area by stretching the surface. Moreover, stretch rates created by such large-scale 42 turbulence can change local flame structure and even extinguish combustion locally. However, the 43 large-scale turbulence can neither penetrate local flames nor directly increase magnitudes of 44 fluctuations of various mixture characteristics (e.g., density, temperature, species mass fractions, or 45 reaction rates) within local flames (i.e., fluctuation magnitudes conditioned to the local values of a 46 combustion progress variable). On the contrary, small-scale turbulent structures, i.e., structures whose 47 length scale is smaller than  $\delta_L$ , could penetrate local flames, thus, intensifying mixing and increasing 48 fluctuation magnitudes inside them. While such small-scale effects are widely expected to exist under 49 certain conditions, there is consensus neither regarding particular manifestations of these effects nor 50 regarding conditions under that such manifestations are of importance.

51 More specifically, in the first combustion regime diagrams, broadening of local flames by small-scale turbulence was hypothesized under conditions of  $\eta_K < \delta_L$ .<sup>13,15</sup> If (i)  $\eta_K = LRe_t^{-3/4}$ , (ii)  $\tau_K = \tau_t Re_t^{-1/2}$ , 52 and (iii)  $\delta_L = v_u/S_L$ , as often assumed, <sup>13-15</sup> a criterion of  $\eta_K < \delta_L$  reads Ka > 1. Here,  $Re_t = u'L/v_u$ 53 54 is turbulent Reynolds number and  $v_u$  is kinematic viscosity of unburned mixture. Later, Peters<sup>21</sup> emphasized that reaction zones could remain thin if  $\delta_r < \eta_K < \delta_L$ , because the reaction zone thickness 55  $\delta_r \ll \delta_L$  within the framework of the classical thermal theory<sup>22</sup> of laminar premixed flames. By 56 assuming that  $\delta_r/\delta_L = 0.1$ , Peters<sup>18,21</sup> suggested a criterion of Ka = 100 to be an upper boundary of 57 58 thin reaction zone regime provided that simplifications (i)-(iii) held. However, since complex-chemistry 59 flames are characterized by significantly larger  $\delta_r/\delta_L$ , with this ratio being as large as 0.5 in moderately 60 lean and near stoichiometric hydrogen-air flames, that boundary should be associated with significantly lower Karlovitz numbers.<sup>23</sup> Nevertheless, the criterion Ka = 100 was widely accepted over almost two 61 62 decades, with both local broadening of preheat zones and existence of thin reaction zones being documented in experimental and Direct Numerical Simulation (DNS) studies, reviewed elsewhere.<sup>24-26</sup> 63 Accordingly, appearance of thickened reaction zones at Ka > 100 was often assumed, but evidence of 64 65 such a regime is still rare.

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66 On the contrary, recent measurements and DNSs put the utility of criteria of both Ka = 1 (broadening 67 of preheat zones) and Ka = 100 (broadening of reaction zones) into question. First, various 68 experimental and DNS data reviewed elsewhere<sup>25,26</sup> indicate that reaction zones are thin even if  $\eta_K$  is 69 much smaller than  $\delta_r$ . In the latest measurements,<sup>27,28</sup> thin reaction zones were documented at *Ka* 70 significantly larger than 100, while broadened reaction zones were also reported at Ka = 590 by Fan 71 et al<sup>28</sup> (while a value of *Ka* is sensitive to its definition,<sup>23</sup> definitions adopted by Peters<sup>18,21</sup> and by Fan 72 et al.<sup>27,28</sup> are consistent).

Second, a recent experimental study by Skiba et al.<sup>29</sup> has shown that a criterion of broadening of flame preheat zones by small-scale turbulence should quantitatively and qualitatively differ from Ka =1, with the influence of turbulent structures smaller than  $\delta_L$  on the internal structure of such zones being weak under conditions of those measurements.

Moreover, there are other data that imply weak influence of small-scale (when compared to  $\delta_L$ ) turbulence on premixed flames. For instance, by running 2D numerical simulations of interactions of a laminar premixed flame and a vortex pair, Poinsot et al.<sup>30</sup> have shown that too small vortices decay rapidly and do not substantially perturb the flame. Subsequent numerical and experimental research into this and similar problems (e.g., interactions of a laminar premixed flame and a single vortex) supported the above conclusion, as reviewed elsewhere,<sup>31</sup> and reported also in recent papers.<sup>32,33</sup>

Besides, Poludnenko and Oran<sup>34</sup> simulated highly turbulent premixed flames by adopting numerical 83 84 meshes with different cell sizes to vary the small-scale branch of turbulence spectrum (in the cited 85 study, kinematic viscosity was set equal to zero and turbulence spectra were bounded by numerical 86 diffusion, which depended directly on the cell size). Reported results did not show substantial 87 perturbations of internal structure of reaction zones even when energy cascade simulated in nonreactive 88 turbulence extended to length scales significantly smaller than  $\delta_r$ . While moderate broadening of flame 89 preheat zones was documented in certain cases, the authors have concluded<sup>34</sup> that "the action of smallscale turbulence is suppressed throughout most of the flame" and "small-scale motions in cold fuel do 90 91 not affect the evolution of the flame brush".

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92 Furthermore, numerical results obtained by Aspden<sup>35</sup> by artificially varying mixture viscosity in a 93 DNS series do not show substantial influence of the viscosity on 2D slices of fuel concentration, 94 temperature, and fuel consumption rate, while vorticity fields simulated in different cases are 95 significantly different. Accordingly, Aspden<sup>35</sup> discussed "suppression of turbulence through the flame" 96 and attributed this effect mainly to "fluid expansion through the flame".

97 In addition, Doan et al.<sup>36</sup> analyzed DNS data to compare contributions of turbulent structures of 98 different scales to flame straining and reported that the studied flames were primarily strained by 99 structures whose length scale was larger than  $2\delta_L$ .

100 Thus, there is a plenty of evidence of inability of turbulent structures whose length scales are smaller 101 than thickness of flame preheat zones to substantially affect the internal structure of such zones and to 102 broaden them. Such findings are often attributed to decay of small-scale turbulence within flame preheat zones<sup>24,26,29</sup> due to (i) the local increase in the temperature and, hence, the mixture viscosity and (ii) an 103 104 increase in the eddy size due to thermal expansion. Both physical mechanisms cause an increase in the 105 length scale of the smallest eddies, thus, resulting in disappearance or weakening of these eddies. The 106 latter physical mechanism may be prioritized based on the aforementioned DNS data by Poludnenko 107 and Oran34 and by Aspden.35

108 While a hypothesis about decay (disappearance or weakening) of the smallest turbulent eddies within 109 flame preheat zones appears to be convincing from qualitative perspective and, therefore, is widely accepted, the present authors are aware of a single study aiming at exploring this hypothesis in turbulent 110 111 flows, where evolution of small-scale turbulence is affected not only by viscous and thermal expansion 112 effects, but also by kinetic energy transfer from larger eddies (such a flux does not appear during 113 interactions of a single vortex or vortex pair with a laminar premixed flame). Wabel et al.<sup>37</sup> performed 114 2D measurements of variations of several turbulence characteristics (kinetic energy, strain rate, and a 115 "conditioned" integral length scale) in the vicinity of reaction zones of highly turbulent flames. The 116 reported results do not show a substantial decay of the turbulent kinetic energy in flame preheat zones but do show an increase in the length scale in such zones. These findings were interpreted to indicate 117 118 decay of small-scale turbulence in flame preheat zones. However, the study did not provide direct

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119 evidence of this decay, because the smallest-scale turbulent eddies could not be resolved using the stateof-the-art optical diagnostic tools under conditions of these measurements. 120

Thus, evolution of small-scale (when compared to  $\delta_L$ ) turbulence in flame preheat zones and 121 122 influence of this turbulence on the inner structure of such zones and their surface area still challenge the combustion community. The present work addresses these fundamental issues by analyzing DNS 123 data obtained earlier by Dave et al.<sup>38,39</sup> from a turbulent, complex-chemistry, lean hydrogen-air flame. 124 125 In the next section, the DNS attributes and applied numerical diagnostic techniques are summarized. 126 Numerical results are reported and discussed in Sec. III, followed by conclusions.

### II. DNS ATTRIBUTES AND PROCESSING METHODS 127

Since the DNS attributes were already reported in earlier papers,<sup>38-43</sup> only a summary is provided 128 129 below. A statistically planar, moderately lean H<sub>2</sub>/air turbulent flame in a box ( $19.18 \times 4.8 \times 4.8$  mm) 130 was simulated invoking a detailed chemical mechanism (21 reactions between 9 species) by Li et al.<sup>44</sup> 131 and the mixture-averaged model of molecular transport. The equivalence ratio, unburned gas 132 temperature, and pressure were set equal to 0.81, 310 K, and 0.1 MPa, respectively. Under such conditions,  $S_L = 1.84$  m/s,  $\delta_L = (T_b - T_u)/\max|\nabla T| = 0.36$  mm, and  $\tau_f = 0.20$  ms. Here, subscripts 133 134 *u* and *b* designate unburned and burned gas, respectively.

135 Three-dimensional unsteady compressible governing equations (i.e., continuity, Navier-Stokes, energy and species transport equations) were numerically solved adopting the Pencil code<sup>45</sup> and a 136 numerical mesh of 960 × 240 × 240 cells. At the inlet and outlet, Navier-Stokes characteristic 137 boundary conditions<sup>46</sup> were set. At four other sides, the boundary conditions were periodic. 138

139 Homogeneous isotropic turbulence was pre-generated using large-scale forcing and fully periodic 140 boundary conditions in a cube. Subsequently, the turbulence evolved until a statistically stationary state 141 with Kolmogorov-Obukhov's 5/3-spectrum was reached.38 During combustion simulations initiated by 142 embedding a planar laminar flame into the computational domain at  $x = x_0$  and t = 0, this turbulence 143 entered the computational domain at a constant mean velocity through the left boundary. The injected 144 turbulence decayed along the x-axis. The flame propagated in the opposite direction against a turbulent 145 flow.

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This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset The pre-generated turbulence was characterized<sup>39</sup> by u' = 6.7 m/s, L = 3.1 mm,  $\eta_K =$ 146  $(v_u^3/\langle \varepsilon \rangle)^{1/4} = 0.018 \text{ mm}, \ \tau_K = (v_u/\langle \varepsilon \rangle)^{1/2} = 0.015 \text{ ms}, \ \text{and} \ Re_t = u'L/v_u = 950. \ \text{Here}, \ \langle \varepsilon \rangle = 0.015 \text{ ms}$ 147  $(2\nu S_{ij}S_{ij})$  is the dissipation rate, averaged over the cube;  $S_{ij} = (\partial u_i / \partial x_j + \partial u_j / \partial x_i)/2$  is the rate-of-148 149 strain tensor; the summation convention applies to repeated indexes. At the leading edge of the mean 150 flame brush, associated here with transverse-averaged temperature-based combustion progress variable  $\bar{c}_T(x) = 0.01$ , the turbulence characteristics were different, i.e., u' = 3.3 m/s, Taylor length scale  $\lambda =$ 151  $\sqrt{10\nu_u\bar{k}/\bar{\varepsilon}} = 0.25$  mm or  $0.69\delta_L$ ,  $\eta_K = 0.018$  mm or  $0.05\delta_L$ , and  $\tau_K = 0.087$  ms. Accordingly, 152  $Re_{\lambda} = u'\lambda/\nu_u = 55, Ka = 2.3$ , while  $(\delta_L/\eta)^2$  is about 400. The difference in Ka and  $(\delta_L/\eta)^2$  is very 153 large, because  $S_L \delta_L / \nu_u \gg 1$  in moderately lean H<sub>2</sub>-air mixtures.<sup>23</sup> 154 PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0153089

155 Average quantities and Probability Density Functions (PDFs) reported in the next section are conditioned to the local values  $c_F = (Y_F - Y_{F,u})/(Y_{F,b} - Y_{F,u})$  of fuel-based combustion progress 156 157 variable, where  $Y_{\rm F}$  designates fuel mass fraction. For instance, the conditioned value  $\langle q | \xi \rangle$  of the 158 quantity q is sampled as follows

$$2\langle q|\xi\rangle(\xi,t) = \iiint q(\mathbf{x},t)I(\mathbf{x},t,\xi)d\mathbf{x},\tag{1}$$

159 where the indicator function  $I(\mathbf{x}, t, \xi) = H[c_F(\mathbf{x}, t) - \xi + \Delta \xi] - H[c_F(\mathbf{x}, t) - \xi - \Delta \xi]$  is equal to unity if  $\xi - \Delta \xi < c_F(\mathbf{x}, t) < \xi + \Delta \xi$  and vanishes otherwise, H designates Heaviside function, and  $0 \le \xi \le$ 160 1 is sampling variable. Similarly, a conditioned PDF  $P(q, \xi)$  is solely yielded by a narrow zone where 161  $I(\mathbf{x}, t, \xi) = 1$  or  $\xi - \Delta \xi < c_F(\mathbf{x}, t) < \xi + \Delta \xi$ . Such zones are very thin, because  $\Delta \xi = 0.005$ , i.e., 100 162 bins are adopted for  $c_F(\mathbf{x}, t)$ , as well as for all other quantities  $q(\mathbf{x}, t)$  whose PDFs are sampled. Thus, 163 data conditioned, e.g., to  $c_F(\mathbf{x}, t) = 0.5$  are sampled from zones where  $0.495 < c_F(\mathbf{x}, t) < 0.505$ . 164 Turbulent fluctuations inside so thin zones are controlled by small-scale turbulent structures. 165

166 Results reported in the following were sampled at 55 instants from 1.291 ms till 1.566 ms.

### III. RESULTS AND DISCUSSION 167

168 Figure 1 reports PDFs of local temperature  $T(\mathbf{x}, t)$ , local mass fraction  $Y_{\rm H}(\mathbf{x}, t)$  of atomic hydrogen,

- 169 and local heat release rate  $\dot{\omega}_T(\mathbf{x}, t)$ , conditioned to the local values of  $c_F(\mathbf{x}, t)$ . Each of these quantities
- 170 is normalized using its value taken at the same  $c_F = c_F(\mathbf{x}, t)$  in the unperturbed (stationary, planar, and

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studied case.

(a)

171 one-dimensional) laminar premixed flame. For instance, the local temperature plotted for  $c_F = 0.5$  is

172 normalized using 1002 K, with the adiabatic combustion temperature being equal to 2177 K in the





(b)

(c)

The sampled conditioned PDFs become narrower with increasing  $c_F(\mathbf{x}, t)$ , at least if  $c_F \leq 0.7$  (this 177 is better seen by comparing the PDF peaks at various  $c_F$ ). This trend indicates a decrease in the range 178 179 (magnitude) of fluctuations of the considered local flame characteristics, with such fluctuations being conditioned to narrow spatial zones and, hence, being controlled by structures whose length scale is 180 181 smaller than the laminar flame thickness. Therefore, the results plotted in Fig. 1 imply reduction of the 182 influence of the small-scale turbulence on the internal structure of local flamelets as the fluid advances from the reactants to the products. This trend is well (less) pronounced for  $Y_{\rm H}({\bf x},t)$  and  $\dot{\omega}_T({\bf x},t)$ 183 184 (temperature, respectively), especially, when  $c_F$  increases from 0.2 to 0.4. Here, different eventual 185 physical mechanisms, e.g., intensification of mixing or change or reaction rates, are not separated, but 186 solely the total influence of turbulence on the PDFs is considered. Since the studied PDFs degenerate to the Dirac delta function  $\delta(x - 1)$  in the laminar flame, the PDF thickness directly characterizes such 187 188 a total influence. Moreover, since conditioned dependencies of  $\langle Y_k | \xi \rangle$ ,  $\langle T | \xi \rangle$ ,  $\langle \rho | \xi \rangle$ , and the rates  $\langle \dot{\omega}_k | \xi \rangle$ 189 of production/consumption of various species on  $\xi$ , sampled from the studied turbulent flame, are close to the counterpart dependencies obtained from the unperturbed laminar flame,<sup>40,41</sup> instantaneous local 190 191 flames may be associated (statistically) with flamelets under conditions of the present study, with the 192 influence of turbulence on the conditioned rates  $\langle \dot{\omega}_k | \xi \rangle$  being statistically weak. Such an association is

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- also supported by sufficiently small widths of PDFs plotted for  $c_F > 0.4$  in Fig. 1. Accordingly, instantaneous local flames are called flamelets henceforth.
- 195 In Fig. 2, a similar trend (PDF constriction with increasing  $c_F$ , which results in increasing the PDF 196 peak with increasing  $c_F$ ) is well pronounced for dilatation  $\Theta \equiv \nabla \cdot \mathbf{u}$  if  $0.3 < c_F < 0.7$  and for flame 197 surface density  $|\nabla c_F|$  if  $0.3 < c_F < 0.8$ . Here,  $|\nabla c_F|$  and  $\Theta$  are normalized using the laminar flame 198 thickness  $\delta_L$  and the dilatation  $\Theta_L = (\sigma - 1)S_L/\delta_L$  in the laminar flame, rather than  $|\nabla c_F|(c_F)$  and  $\nabla \cdot$ 199  $\mathbf{u}(c_F)$  in the laminar flame;  $\sigma = \rho_u/\rho_b$  is the density ratio.



- Fig. 2. Probability density functions for normalized (a) flame surface density  $\delta_L |\nabla c_F(\mathbf{x}, t)|$  and (b) dilatation  $\Theta_L^{-1} \nabla \cdot \mathbf{u}(\mathbf{x}, t)$ , conditioned to the local values of  $c_F(\mathbf{x}, t)$  specified in legends and sampled from the entire computational domain at 55 instants.
- The results reported in Figs. 1 and 2 are fully consistent with weak influence of turbulence on local structure of reaction zones, highlighted in Sect. 1. However, Figs. 3 and 4 do not allow us to attribute
- this weak influence to the local turbulence decay within flamelets.





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Indeed, first, Fig. 3a shows that the normalized conditioned rms velocities  $(\langle u_i^2 | \xi \rangle - \langle u_i | \xi \rangle^2)^{1/2} / S_L$ , 208 see broken lines, as well as  $\langle u'|\xi\rangle/S_L \equiv \left(\sum_{i=1}^3 \left[\langle u_i^2|\xi\rangle - \langle u_i|\xi\rangle^2\right]/3\right)^{1/2}/S_L$ , see solid line, do not 209 decrease with increasing  $\xi$ . Contrary, the observed trend is opposite at  $\xi < 0.7$ , while weakly 210 211 pronounced. The former numerical result is consistent with experimental data by Wabel et al.37 who did 212 not document a decrease in the conditioned turbulent kinetic energy with a combustion progress variable 213 either. The sole effect of combustion on the rms velocities, observed in Fig. 3a, consists of anisotropy 214 of the fluctuating velocity field due to acceleration of the flow in the axial direction, which local flames are predominantly normal to under the studied conditions.43 215

Second, Figs. 3b and 3c indicate that the normalized flame strain rate  $\tau_f \langle a_t | \xi \rangle$ , see Fig. 3b, the 216 normalized total rate of strain  $\tau_f^2 \langle S^2 | \xi \rangle$ , enstrophy  $\tau_f^2 \langle \omega^2 | \xi \rangle$ , and dissipation rate  $v_u^{-1} \tau_f^2 \langle \varepsilon | \xi \rangle$ , see red 217 218 solid, black dotted-dashed, and blue dashed lines, respectively, in Fig. 3c, do not show a substantial decrease with  $\xi$  if  $\xi < 0.8$ . Here,  $a_t = \nabla \cdot \mathbf{u} - \mathbf{n}(\nabla \mathbf{u})\mathbf{n}$ ,  $S^2 = S_{ij}S_{ij}$ ,  $\omega^2 = \omega_i\omega_i$ , and  $\varepsilon = \omega_i\omega_i$ 219  $2\nu(S^2 - \Theta^2/3)$ ;  $\mathbf{n} = -\nabla c_F/|\nabla c_F|$  is the unit vector locally normal to flame surface and pointing to 220 221 unburned reactants; and  $\boldsymbol{\omega} = \nabla \times \mathbf{u}$  is vorticity vector. While the conditioned rate of strain decreases with  $\xi$  at  $\xi > 0.3$ , the opposite and more pronounced trend is observed at small  $\xi$  and  $\tau_{\ell}^2 \langle S^2 | \xi = 0.8 \rangle$ 222 is still larger than  $\tau_f^2 \langle S^2 | \xi = 0.05 \rangle$ . 223

224 The considered conditioned small-scale turbulence and flame characteristics decrease rapidly with 225 increasing  $\xi$  at  $\xi(\mathbf{x}, t) > 0.8$ . Such large values of  $\xi$  are associated with a radical recombination zone, where the temperature is gradually increases due to relatively low heat release in three-molecular 226 recombination reactions.<sup>47</sup> Due to a relatively large thickness of this zone, larger-scale turbulent eddies 227 can penetrate it and perturb its structure, e.g., PDFs plotted in Figs. 1a, 1c, or 2a are wider at  $\xi = 0.8$ 228 229 than at  $\xi = 0.7$ . However, the influence of a larger scale turbulent structures on the radical 230 recombination zone is beyond the scope of the present work, whose focus is placed on preheat and 231 reaction zones of instantaneous local flames.

Sufficiently weak  $\xi$ -dependencies of  $\tau_f^2 \langle S^2 | \xi \rangle$  and  $\tau_f^2 \langle \omega^2 | \xi \rangle$ , sampled from the DNS data within flamelets, are also consistent with the experimental data by Wabel et al.,<sup>37</sup> who reported 2D counterparts

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of  $S^2$  and  $\omega^2$ . However, those experimental data characterized moderately small velocity gradients resolved in the measurements, whereas  $\tau_f^2 \langle S^2 | \xi \rangle$  and  $\tau_f^2 \langle \omega^2 | \xi \rangle$  sampled from the present DNS data appear to be controlled by the smallest turbulent structures of the Kolmogorov scale, which is much less than  $\delta_L$  under conditions of the analyzed DNS.

Third, Fig. 4 also shows very weak variations of strain rate  $a_t$  within flamelets (PDFs conditioned to different  $\xi$  are barely distinguishable in Fig. 4a); a weak decrease (increase) in fluctuations of  $S^2$  with increasing  $\xi$  if  $\xi > 0.3$  ( $\xi < 0.3$ , respectively), as the PDFs in Fig. 4b become narrower (wider, respectively); and an increase in fluctuations of  $\omega^2$  with increasing  $c_F$  (the PDFs become wider in Fig. 4c), which is most pronounced at  $\xi < 0.4$ .



**Fig. 4.** Probability density functions for normalized (a) strain rate  $\tau_f a_t$ , (b) rate of strain  $\tau_f^2 S^2$ , and (c) enstrophy  $\tau_f^2 \omega^2$ , conditioned to the local values of  $c_F(\mathbf{x}, t)$  specified in legends and sampled from the entire computational domain at 55 instants.

245 Thus, decay of small-scale turbulence within flamelets is indicated neither in Fig. 3 nor in Fig. 4. Accordingly, the constriction of PDFs of local combustion characteristics (i.e., a decrease in the 246 247 magnitude of fluctuations of these characteristics) with increasing  $\xi$ , observed at 0.3 <  $\xi$  < 0.7 in Figs. 248 1 and 2, should not be attributed to decay of small-scale turbulence within flamelets, as often believed. 249 Therefore, to resolve this conundrum and to make Figs. 1 and 2 consistent with Figs. 3 and 4, another 250 cause of reduction of the influence of small-scale turbulence on the local flamelet structure, shown in 251 Figs. 1 and 2, should be revealed. Rapid flow acceleration, mentioned earlier by Poludnenko and Oran,<sup>34</sup> 252 appears to be a relevant physical mechanism. Indeed, if following activation energy asymptotic theories of stretched laminar flames,48,49 we 253

- 254 consider strain rate  $a_t$  to be the most appropriate quantity for characterizing the influence of turbulence
- 255 on premixed flame structure, Fig. 3b shows that a product of  $\tau_f \langle a_t | \xi \rangle$  is about (but less than) two.

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However, due to rapid acceleration of the flow, caused by combustion-induced thermal expansion, the residence time  $\tau_r(0.3 < c_F < 0.7)$  of turbulent structures within flamelet zones characterized by 0.3 <  $c_F < 0.7$  should be substantially shorter than  $\tau_f$ . In the following, if the opposite is not stated, this residence time will be designated with symbol  $\tau_r$  without parentheses for brevity. Therefore, a product of  $\tau_r \langle a_t | c_F \rangle$  is expected to be less than unity so that the turbulence does not have time enough to significantly perturb the local flamelet structure at  $0.3 < c_F < 0.7$ .



Figure 5. (a) Variations in the density in the unperturbed laminar flame (black solid line) and the conditioned density  $\langle \rho | \xi \rangle$  in the turbulent flame (red dashed line). (b) Normalized conditioned derivative  $\tau_f \langle \mathbf{n} \cdot \nabla (\mathbf{u} \cdot \mathbf{n}) | \xi \rangle$  of the locally normal velocity **u** · **n**, taken along the local flamelet normal. (c) Normalized conditioned flame surface density  $\delta_L \langle | \nabla c_F | | \xi \rangle$ . (d) Normalized residence time  $\tau_r (c_F) / \tau_f$  in turbulent flamelet (red solid line) and laminar flame (black dashed line) zones bounded by  $c_F -$ 0.005 and  $c_F + 0.005$ .

Density variations reported in Fig. 5a seem to indirectly support this hypothesis. Indeed, Fig. 5a shows that the local density is decreased by a factor of two already at  $c_F = 0.2$  or by a factor of larger than three at  $c_F = 0.6$ . Consequently, the local normal flow velocity with respect to flamelets is increased (when compared to  $S_L$ ) by a factor of two at  $c_F = 0.2$  or by a factor of larger than three at  $c_F = 0.6$ . Under conditions of the present DNS, differences in density-weighted displacement speed  $S_d^* = \rho S_d / \rho_u$  and  $S_L$  are statistically weak<sup>39</sup> and flamelets statistically retain the unperturbed laminar

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flame structure,<sup>40,41</sup> see Fig. 5a also, thus, supporting the use of the velocity  $\rho_u S_L / \rho$  for estimating the residence time. It is also worth noting that the dependence of  $\langle \rho | \xi \rangle$  on  $\xi$ , plotted in Fig. 5a, is significantly steeper at  $\xi < 0.3$  than at larger  $\xi$ . Accordingly, by virtue of  $S_d \approx \rho_u S_L / \rho$ , the local increase in  $S_d$  with increasing  $c_F$  should be more pronounced (statistically) at  $c_F < 0.3$ . Therefore, the rate of reduction of the influence of turbulence on the local flamelet structure should be higher (statistically) at low  $c_F$ . Indeed, such a trend is observed for the rate  $\dot{\omega}_T(\mathbf{x}, t)$  in Fig. 1c and, especially, for  $Y_H(\mathbf{x}, t)$  in Fig. 1b.

Significant acceleration of the locally normal flow in flamelet zones characterized by  $0.2 < c_F < 0.7$ is further supported in Fig. 5b, which shows the normalized conditioned derivative  $\tau_f \langle \mathbf{n} \cdot \nabla (\mathbf{u} \cdot \mathbf{n}) | \xi \rangle$ of the locally normal velocity  $\mathbf{u} \cdot \mathbf{n}$ , taken along the local flamelet normal  $\mathbf{n}$ . Here, the negative derivative indicates acceleration of the flow in the direction opposite to  $\mathbf{n}$ , i.e., to products. Due to the rapid acceleration of the locally normal flow, the residence time of turbulent fluid within the discussed flamelet zone ( $0.2 < c_F < 0.7$ ) should be reduced. In addition, the residence is relatively short due to a small thickness of this zone due to large local values of  $|\nabla c_F|$ , see Fig. 5c.

The joint effect of these two factors (large local  $|\langle \mathbf{n} \cdot \nabla (\mathbf{u} \cdot \mathbf{n}) | \xi \rangle|$  and large local  $|\nabla c_F|$ ) is illustrated in Fig. 5d, which reports variations of the normalized residence time  $\tau_r(c_F)/\tau_f$  in the laminar flame zones bounded by  $c_F \pm \Delta c_F$  where  $\Delta c_F = 0.005$ , see black solid line. This normalized time, evaluated as follows

$$\frac{\tau_r(c_F)}{\tau_f} = \frac{1}{\tau_f u(x)} \frac{\Delta c_F}{|dc_F/dx|'}$$
(2)

is much less than unity, partially because it characterizes thin zones associated with  $\Delta c_F = 0.01$ . However, even the residence time  $\tau_r$  in a significantly thicker zone of  $0.195 < c_F < 0.705$ , which has been estimated to be a sum of residence times evaluated using Eq. (2) for  $c_F = 0.2 \pm \Delta c_F$ ,  $c_F = 0.21 \pm \Delta c_F$ , ..., and  $c_F = 0.7 \pm \Delta c_F$ , is equal to  $0.18\tau_f$ , whereas the Kolmogorov time scale  $\tau_K$  evaluated at the leading edge of the mean flame brush is larger by a factor of 2.5. This comparison of  $\tau_r$  and  $\tau_K$ supports a hypothesis that the residence time is too short and even the smallest-scale turbulent fluctuations creating the largest velocity gradients do not have enough time to significantly perturb the

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local flamelet structure at  $0.2 < c_F < 0.7$ . When  $c_F$  tends to zero or unity, the local residence time rapidly grows. For instance, it is equal to  $0.42\tau_f$  and  $1.07\tau_f$  for  $0.02 < c_F < 0.2$  and  $0.7 < c_F < 0.98$ , respectively. Accordingly, the influence of small-scale turbulence on local flamelet structure is more pronounced at low and large  $c_F$ .

While black dashed line in Fig. 5d shows results obtained from the laminar flame, these results characterize the residence time within turbulent flamelets to the leading order, because the flamelet structure is weakly perturbed from the statistical perspective under conditions of the present DNS study, as shown earlier,<sup>40,41</sup> see Fig. 5a also. In addition, the following simplified estimate

$$\frac{\tau_r(\xi)}{\tau_f} = \frac{1}{\tau_f u(n)} \frac{\Delta \xi}{\langle |\nabla c_F||\xi \rangle}, \qquad u(n) = S_L - \int_{c_*^*}^{\varsigma} \langle \mathbf{n} \cdot \nabla (\mathbf{u} \cdot \mathbf{n}) |\zeta \rangle \frac{d\zeta}{\langle |\nabla c_F||\zeta \rangle}, \tag{3}$$

ξ

306 of the normalized residence time within turbulent flamelets was also performed. Here, when compared 307 to Eq. (2), (i) distance along the locally normal (to the flamelet) direction is estimated to be equal to 308  $\Delta \xi / \langle | \nabla c_F | | \xi \rangle$ , (ii) flow velocity in the laminar flow is substituted with the locally normal flow velocity 309 u(n), (iii) which is estimated by integrating the locally normal velocity gradient  $\langle \mathbf{n} \cdot \nabla (\mathbf{u} \cdot \mathbf{n}) | \zeta \rangle$  along the normal direction, with (iv) the integration being performed starting from a value of  $\xi$ , associated 310 311 with the change of the sign of  $\langle \mathbf{n} \cdot \nabla (\mathbf{u} \cdot \mathbf{n}) | \xi \rangle$  from positive at  $\xi < c_F^*$  (in such flamelet zones, turbulent 312 fluctuations of velocity statistically overwhelm velocity changes due to the local density variations) to 313 negative at  $\xi > c_F^*$  (in such flamelet zones, local acceleration of the normal flow is mainly controlled 314 by heat release). Due to the significant influence of turbulent velocity fluctuations on  $\langle \mathbf{n} \cdot \nabla (\mathbf{u} \cdot \mathbf{n}) | \xi \rangle$ 315 at low  $\xi$ , the choice of the integration boundary  $c_F^*$  is not rigorous. Accordingly, a correlation between velocity and scalar gradients is also neglected in Eq. (2), i.e., the conditioned ratio 316  $\langle \mathbf{n} \cdot \nabla (\mathbf{u} \cdot \mathbf{n}) / | \nabla c_F | | \xi \rangle$  is substituted with the ratio  $\langle \mathbf{n} \cdot \nabla (\mathbf{u} \cdot \mathbf{n}) | \xi \rangle / \langle | \nabla c_F | | \xi \rangle$  of conditioned quantities. 317 Nevertheless, the residence times calculated using Eqs. (1) and (2) are close to one another at 0.2 <318 319  $\xi < 0.7$ , thus, further supporting the residence-time hypothesis.

320 This hypothesis could also be useful for understanding the lack of substantial decay of various 321 turbulence characteristics as turbulent structures move from  $c_F = 0.2$  to  $c_F = 0.7$  within flamelets, see 322 Figs. 3 and 4. For instance, the residence time could be insufficient for the dissipation of turbulence due

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to molecular viscosity to substantially decrease magnitudes of local velocity gradients. If we (i) estimate 323 time required for this dissipation as follows  $\tau_{\varepsilon} = (\nu/\varepsilon)^{1/2}$ , (ii) take the highest value of the conditioned 324 dissipation rate  $v_u^{-1}\tau_f^2\langle\varepsilon|\xi\rangle = 93$ , reported in Fig. 3c, and (iii) allow for an increase in the viscosity v325 at  $\xi = 0.6$  or T = 1130 K, associated with this highest value, i.e.,  $v(c_F = 0.6) = 2.5v_u$ ; we obtain the 326 dissipation time scale that is shorter by a factor of 2.7 than the Kolmogorov time scale  $\tau_K$  evaluated at 327 328 the leading edge of the mean flame brush. Therefore, the dissipation time scale can be as low as  $0.9\tau_r$ 329 and magnitude of velocity gradients could be decreased by viscous dissipation, e.g., see red solid line in Fig. 3c (at  $\xi > 0.3$ ) or PDFs reported in Fig. 4b. However, since even the shortest dissipation time 330 scale is comparable with the residence time  $\tau_r$ , the observed decrease in  $\tau_f^2(S^2|\xi)$  with  $\xi$  is sufficiently 331 slow. Moreover, the influence of combustion-induced thermal expansion on a turbulent flow is not 332 reduced to an increase in the kinematic viscosity with the temperature and other physical mechanisms 333 discussed in detail elsewhere<sup>50,51</sup> can also play an important role. For instance, generation of vorticity 334 by baroclinic torque, appears to control an increase in (i) the conditioned enstrophy  $\tau_f^2(\omega^2|\xi)$  at 0.2 < 335  $\xi < 0.8$ , see black dotted-dashed line in Fig. 3c, or (ii) magnitude of its fluctuations, see Fig. 4c. 336 337 Besides, generation of potential velocity gradients and reduction of vorticity due to dilatation appear to control an increase  $\tau_{\ell}^2 \langle S^2 | \xi \rangle$  at  $0.05 < \xi < 0.3$  and a decrease in  $\tau_{\ell}^2 \langle \omega^2 | \xi \rangle$  at  $\xi < 0.15$ , respectively, 338 339 see Fig. 3c. An analysis of these physical mechanisms will be a subject for future work but is beyond 340 the scope of the present study. Nevertheless, the limited residence time of small-scale turbulence within 341 flamelets could also play a role and should also be borne in mind.

### 342 IV. CONCLUDING REMARKS

DNS data analyzed in the present work show that (i) the influence of small-scale (when compared to the laminar flame thickness) turbulence on internal flamelet structure is reduced during advection from unburned reactants to combustion products, but (ii) neither the local turbulence characteristics (conditioned rms velocities, total strain, and enstrophy) nor flame strain rate decrease substantially from the reactant side to the product side. To reconcile these two apparently inconsistent trends, the former is hypothesized to stem from shortening of the residence time of the small-scale turbulence within

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flamelets due to combustion-induced acceleration of the local flow in the flamelet-normal direction.Certain DNS data discussed above indirectly support this hypothesis.

351 It is worth recalling that an apparently similar hypothesis highlighting insufficient residence time of 352 turbulence in a flame brush was earlier put forward<sup>52</sup> to explain weak influence of turbulence on the 353 surface of an unburned-mixture-finger.<sup>53</sup> However, the residence time considered in the cited studies is 354 controlled by turbulent motion in unburned reactants from the leading edge of a thick mean flame brush 355 to its trailing edge, whereas the residence time emphasized in the present work is controlled by 356 advection of turbulence through thin flamelets.

357 Finally, it should be stressed that the present study does not indicate that the hypothesized residence-358 time mechanism always dominates, whereas a widely accepted turbulence-decay mechanism is of minor importance. Both physical mechanisms are expected to play an important role, but, likely, at different 359 360 conditions. This issue requires further study, e.g., to find a criterion that separates domains of primarily 361 importance of each mechanism. Since  $S_L \delta_L / v_u$  and, hence, difference between  $Ka = \tau_f / \tau_K$  and  $(\delta_L/\eta_K)^2$  is much larger in lean or near-stoichiometric hydrogen-air flames than in hydrocarbon-air 362 ones, conditions of the present study, i.e., Ka = O(1) despite  $(\delta_L/\eta_K)^2 \gg 1$ , are specific to the former 363 364 flames and are beneficial for the residence-time mechanism. In hydrocarbon-air flames characterized 365 by  $(\delta_L/\eta_K)^2 \gg 1$ , the Karlovitz number Ka should also be much larger than unity and a decrease in the residence time of small-scale turbulence within flamelets is unlikely to make the residence time 366 shorter than  $\tau_K$ . Nevertheless, the hypothesized residence-time mechanism should be borne in mind, at 367 least for hydrogen-air turbulent premixed flames. 368

### 369 Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationshipsthat could have appeared to influence the work reported in this paper.

### 372 Data availability

373 The data that support the findings of this study are available from the corresponding author upon

374 reasonable request.

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