Facilitated Ignition in Turbulence through Differential Diffusion

Fujia Wu,[1] Abhishek Saha,[1] Swetaprovo Chaudhuri,[1] and Chung K. Law[1,2,*]
[1]Department of Mechanical and Aerospace Engineering, Princeton University, Princeton, New Jersey 08544, USA
[2]Center for Combustion Energy, Tsinghua University, Beijing 100084, China
(Received 29 November 2013; revised manuscript received 28 April 2014; published 9 July 2014)

Contrary to the belief that ignition of a combustible mixture by a high-energy kernel is more difficult in turbulence than in quiescence because of the increased dissipation rate of the deposited energy, we experimentally demonstrate that it can actually be facilitated by turbulence for mixtures whose thermal diffusivity sufficiently exceeds its mass diffusivity. In such cases, turbulence breaks the otherwise single spherical flame of positive curvature, and hence positive aerodynamics stretch, into a multitude of wrinkled flamelets subjected to either positive or negative stretch, such that the intensified burning of the latter constitutes local sources to facilitate ignition.

DOI: 10.1103/PhysRevLett.113.024503 PACS numbers: 47.70.Pq, 47.27.-i, 51.20.+d

Ignition of a combustible mixture by a stimulus kernel is of relevance to many terrestrial and extraterrestrial phenomena and applications, such as the various modes of engine operation [1,2], prevention of accidental explosions [3], and initiation of the supernova explosion [4–7]. Frequently, such ignition occurs in flows that are highly turbulent, involving a wide range of time and length scales. Previous studies have advocated that turbulence renders ignition more difficult [8–14], based on the notion that turbulence increases the dissipation rate of the deposited kernel energy before an embryonic flame either has the time or is aerodynamically favorable to develop.

Such an argument, however, does not take into consideration the evolution and dynamics of the structure of the nascent flame kernel after it is formed. To appreciate these influences, and to facilitate exposition of the rationale of the experimental investigation to be presented later, it is first noted that the burning rate and consequently the extinction propensity of a laminar flame segment of surface area A, subjected to generalized aerodynamic stretching which accounts for the collective effects of flow nonuniformity, flame curvature, and flame unsteadiness, is characterized by the stretch rate $K = d(\ln A)/dt$ [15]. Analysis in the limit of large activation energy E_a for an assumed one-step overall reaction typically yields a relation [15–19] having the functional form:

$$s^2 \ln s^2 = -\alpha\kappa, \tag{1}$$

where s is the local, stretched flame speed scaled by the steady unstretched flame speed, κ is the stretch rate K scaled by the laminar flame time, and α is commonly referred to as the Markstein number, which indicates the sensitivity of the flame responses, such as the propagation speed, to stretch. Various theoretical expressions for α have been derived [20–25], accounting for effects of diffusive transport and thermal expansion [20,21], the temperature dependence of the transport properties [22], mixture composition, general reaction orders [23,24], and recently a modeled two-step kinetic scheme [25]. These results show that, depending on the definition of the flame location [23] and specification of the chemical kinetics [25], the specific form for α can be different for flame curvature and flow straining. The dominant influence, however, is the extent that the mixture's effective Lewis number (Le) deviates from unity, as quantified by the parameter (Le − 1), where Le is defined as the ratio of its thermal diffusivity to the controlling mass diffusivity. This is demonstrated, for example, by the expression [24]

$$\alpha = \frac{\sigma}{(\sigma-1)}\int_1^{\sigma}\frac{\lambda(x)}{x}dx + \frac{\mathrm{Ze}(\mathrm{Le}-1)}{2(\sigma-1)}\int_1^{\sigma}\frac{\lambda(x)}{x}\ln\left(\frac{\sigma-1}{x-1}\right)dx, \tag{2}$$

where σ is the thermal expansion ratio across the flame, $\lambda(x)$ the normalized thermal conductivity as a function of the normalized temperature x, $\mathrm{Ze} = E_a(T_b - T_u)/RT_b{}^2$ the Zel'dovich number, R the universal gas constant, and T_b and T_u the burned and unburned gas temperatures, respectively.

Two observations can be made from the above theoretical results. First, by examining the linearized form of Eq. (1), $s \approx 1 - \alpha\kappa$, it is apparent that the trend of the flame response is qualitatively affected by the sign of a combined, diffusivity-affected stretch term, $\Lambda = (\mathrm{Le}-1)\kappa$, weakened for $\Lambda > 0$, and strengthened otherwise. Second, as a consequence of the nonlinear feedback between transport and chemical heat release [18], Eq. (1) exhibits a dual-solution response for $\Lambda > 0$, indicating the potential of extinction at the associated turning point.

Applying the above concept to the sustenance of a spark-ignited, continuously expanding spherical flame in quiescence, it is first recognized that since the stretch rate is given by $K = 2d(\ln R)/dt > 0$, the flame burning rate is governed by the sign of $(\mathrm{Le} - 1)$. It is then obvious that, if enough ignition energy is supplied to establish a flame kernel, then a $\mathrm{Le} < 1$ flame would be strengthened by stretch and thus continuously propagate, albeit with progressively reduced velocity, until it reaches the adiabatic planar flame limit as $R \to \infty$. However, for a $\mathrm{Le} > 1$ flame, since the burning rate is weakened by stretch, even it can be initially established, the ignition energy may not be sufficient to sustain flame propagation when all this energy is dissipated before R has reached a critical flame radius R_c, leading to extinction. Thus, if the ignition energy is slightly larger than that barely sufficient to sustain flame propagation at R_c, then the propagation velocity will exhibit a minimum at R_c and increase thereafter to attain the adiabatic planar flame limit as $R \to \infty$. Consequently, we expect the flame speed to be small at the state of R_c. Indeed, unsteady analyses and simulations of the initiation of the spherical flame [26–28] have shown that R_c corresponds well to the Zel'dovich flame ball radius R_Z, with a vanishing propagation flame speed [29].

The above distinctively different behaviors are shown, respectively, in Figs. 1(a) and 1(b), for the instantaneous propagation velocity $dR(t)/dt$, for lean and rich H_2/air

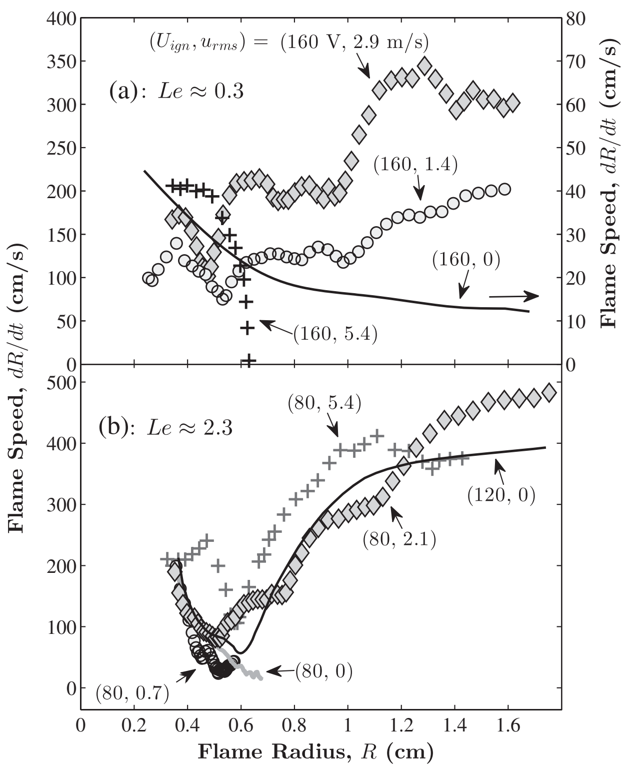

FIG. 1. Flame speed versus flame radius for (a) lean H_2/air at $\phi = 0.18$ and (b) rich H_2/air at $\phi = 5.1$, at different ignition voltages and turbulent levels. The flame radius in turbulent cases is defined as $R = \sqrt{\bar{A}/\pi}$, where $\bar{A}$ is the area enclosed by the flame edge tracked from the Schlieren image. The spark gap distance d_g is 0.30 mm for (a) and 0.58 mm for (b).

mixtures whose Le's are respectively < 1 and > 1; the experimental specifications are given later when presenting the results in turbulence. It is seen from the solid lines that, for the quiescent situation of no turbulence, $u_{\mathrm{rms}} = 0$, an $\mathrm{Le}(= 0.3) < 1$ flame will continuously propagate, with monotonically decreasing velocity, as long as sufficient ignition energy, varied by the spark discharge voltage U_{ign}, is supplied. However, an $\mathrm{Le}(= 2.3) > 1$ flame will extinguish if the ignition energy is not sufficient ($U_{\mathrm{ign}} = 80$ V), but with sufficient ignition energy ($U_{\mathrm{ign}} = 120$ V) will continuously propagate after having attained a minimum, critical radius.

Let us now consider ignition in turbulence, and for the sake of clarity focus on the critical state at which the ignition energy is just barely sufficient to initiate an expanding $\mathrm{Le} > 1$ flame in quiescence. It is clear that, in the presence of turbulence, the otherwise spherical flame with a uniform, positive, curvature will be wrinkled, leading to an embryonic flame structure with both locally positive and negative curvatures. Furthermore, local flows with extensive and compressive strain rates, with $K > 0$ and < 0, can also be induced. The net effect is that the flame surface will be locally subjected to additional $\Lambda > 0$ and < 0 stretch effects. The key point to note here is that since the flame in quiescence is already at the incipient state of extinction over its entire surface, any flame segments that are subjected to additional $\Lambda > 0$ effect in turbulence are still susceptible to extinction. However, the burning intensity of the flame segments that are subjected to $\Lambda < 0$ effect will be increased, moving them away from incipient extinction and consequently can collectively serve as local ignition sources to sustain the global flame structure.

With the above expository anticipation, the present work then aims to explore such a possibility, which may alter the traditional view on the criteria of ignition in turbulent flows, and as such highlight the necessity to incorporate the dynamics of the embryonic flamelet structure into the description and prediction of ignition.

We undertook a well-controlled experimental approach to categorically study the effects of turbulence on an ignition kernel. Tests were conducted in a constant-pressure, optically accessible, vessel, detailed in Ref. [30]. Near-isotropic turbulence is generated by four orthogonally positioned fans and then characterized by high-speed particle image velocimetry. The ignition system was similar to those used in automotive engines, with a slight modification by replacing the spark plug by two tungsten wires of 250 μm diameter and centered axially. The spark is generated by discharging a 33 μF capacitor through an ignition coil with a 134:1 turn ratio. Voltage (U_{ign}) across the capacitor was made variable from 50 to 170 V, allowing the ignition energy to vary.

In designing the matrix of the experimental investigation, it was noted that previous works on the interaction of turbulence and ignition kernel were limited to inhibited ignition, ostensibly due to the small deviations in Le from unity. Therefore, we extended the experiments to cover a wide range of Le, especially focusing on mixtures with Le sufficiently greater than unity such that ignition could be limited by the extent of stretch that the flame kernel experiences. At the same time we also considered mixtures with Le ≈ 1 or Le < 1 to provide a complete description of the phenomena. We selected H_2/O_2 mixtures for most of the investigation because it allows flexible variation of Le due to the drastic difference between the molecular weights of hydrogen and oxygen. This can be achieved by varying the H_2/O_2 ratio, namely, the equivalence ratio ϕ of the mixture (defined to be the fuel/oxygen ratio relative to the stoichiometric mixture), thereby changing the diffusivity of the deficient, hence controlling, reactant. Furthermore, since the H_2/O_2 oxidative chemistry is relatively simple and also reasonably well established, the present data can be usefully adopted in further computational studies on issues of turbulence-chemistry interaction in general and of this practically relevant problem in particular.

We first demonstrate the possible enhancement of ignition by turbulence through direct, time-resolved Schlieren images, using a fuel-rich H_2/air mixture of $\phi = 5.1$, which has a large Le of 2.3 since the controlling, deficient reactant is O_2, while the thermal diffusivity is controlled by that of H_2. Figures 2(a) and 2(b) show, respectively, a successful and a failed ignition event in quiescence ($u_{\rm rms} = 0$), with an ignition voltage, $U_{\rm ign} =$ 120 V and a smaller value of 80 V. This result therefore agrees with the earlier discussion on the dependence of ignition on the energy input. The measured velocities of these images, together with those for the lean mixture of $\phi = 0.18$, with Le $= 0.3$, are those shown in Figs. 1(b) and 1(a), respectively.

If we next maintain the ignition voltage constant at 80 V but progressively increase the turbulence level, Figs. 2(b)–2(e) show that the structure of the flame kernel is changed from a positively stretched spherical surface to a multitude of wrinkled flamelets of both positive and negative curvatures, and that ignition is achieved at $u_{\rm rms} = 2.9$ m/s. This result therefore supports the notion that, for Le > 1, a combustible mixture can be ignited in turbulence with an ignition energy that is not enough to ignite the same mixture in quiescence.

We have extensively mapped out the ignition boundary for the $\phi = 5.1$ mixture in terms of the turbulence intensity and the ignition energy, summarized in Fig. 3(a). It is found that the minimum discharge voltage required for successful ignition in the presence of moderate turbulence ($u_{\rm rms} =$ 2.9 m/s) is 65 V (indicated by blue circles filled with green

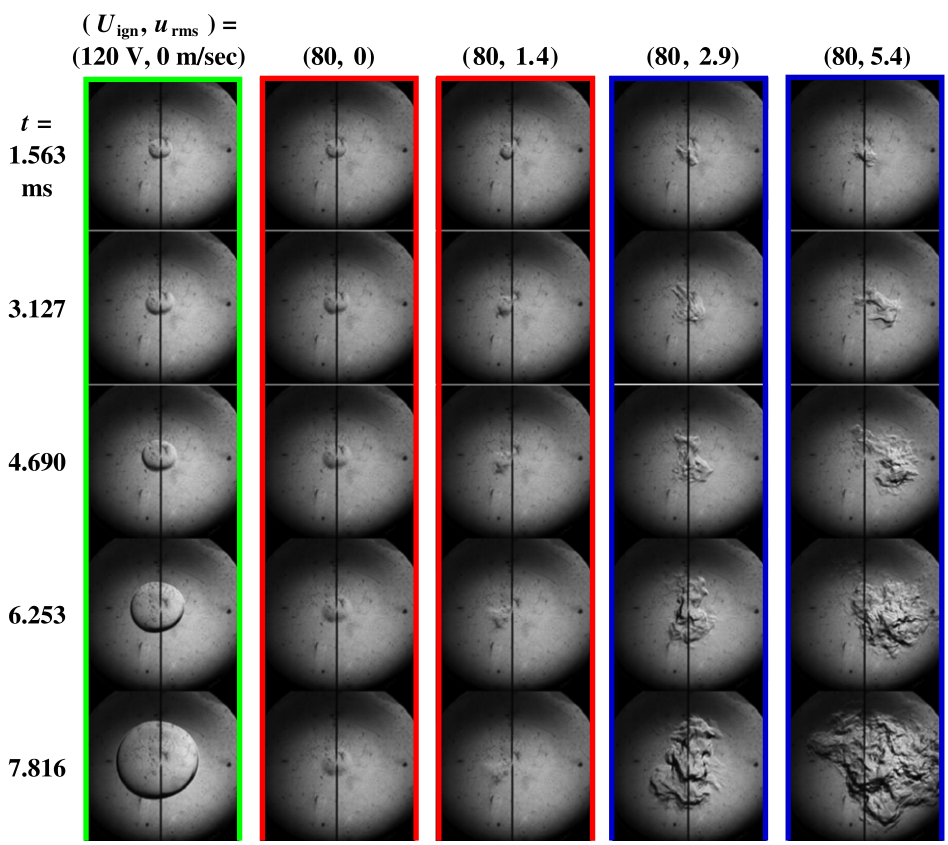

FIG. 2 (color online). Sequential Schlieren images of flame kernel development for H_2/air at $\phi = 5.1$ (Le ≈ 2.3), at different ignition voltages and turbulent levels. Schlieren imaging signal is proportional to the density gradient of the flow field, a good indicator of the flame front. The view for each image is 65 mm $\times$ 65 mm. t is time after discharge.

color in the figure), which is substantially reduced from the voltage required for ignition in quiescence (100 V). Based on the relation $E_{ign} = CU_{ign}^2$, where E_{ign} and C are the discharge energy and capacitance, respectively, this result implies that the minimum ignition energy can be lowered by a few factors with the presence of turbulence.

Having demonstrated the facilitating effect of turbulence on the Le > 1, rich H_2/air mixtures, it is necessary to investigate the response of lean mixtures, whose Le is less than unity because the controlling, deficient reactant is now H_2 and the thermal diffusivity is dominated by those of O_2 and N_2. As anticipated earlier, since a flame kernel with Le < 1 is always facilitated by positive stretch in quiescence, ignition is expected to be successful as long as a flame kernel can be established. Turbulence, in this case, increases the dissipation rate of the deposited kernel energy and as such renders ignition progressively more difficult with increasing turbulence intensity, eventually leading to extinction. Our results on lean H_2/air mixtures [$\phi = 0.12$–0.2, Figs. 3(b) and 3(c)] confirm this mechanistic interpretation and are also consistent with the conclusions of previous studies which, as noted earlier, were mostly based on Le ≈ 1 mixtures.

We also need to rule out the possibility that the controlling ignition chemistry could be different for fuel-rich and fuel-lean mixtures, causing the observed phenomena. To remove such a possibility, we manipulated the values of Le by changing the inert bath gas from N_2 to (He, Ar, CO_2), with the fuel/oxygen ratio fixed such that the controlling lean versus rich chemistry is not affected. It is seen [Figs. 3(d)–3(h)] that the facilitating effect of turbulence is again manifested only for Le > 1 mixtures, and that the effect can be flipped from inhibiting to facilitating solely by changing the inert bath gas while ϕ is fixed at

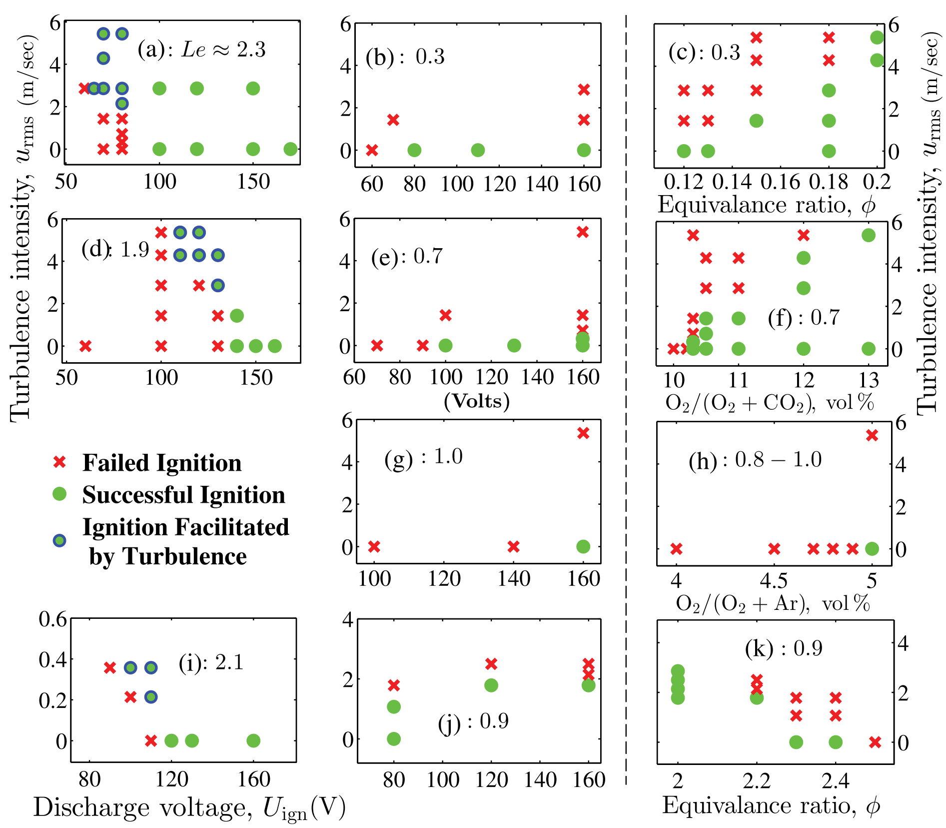

FIG. 3 (color online). Ignition test results plotted against turbulence intensity (u_{rms}) and discharge voltage (U_{ign}) or mixture reactivity, represented by the H_2/O_2 ratio or the amount of inert gas. (a) H_2/air at equivalence ratio (ϕ) = 5.1, (b) H_2/air at $\phi = 0.12$, (c) H_2/air for fixed $U_{ign} = 160$ V. (d) H_2/O_2/He at $\phi = 1.0$, $O_2/(O_2 + \mathrm{He})$ vol % = 8.0%, (e) $H_2/O_2/CO_2$ at $\phi = 1.0$, $O_2/(O_2 + CO_2)$ % vol = 10.3%, (f) $H_2/O_2/CO_2$ at $\phi = 1.0$ for fixed $U_{ign} = 160$ V, (g) H_2/O_2/Ar at $\phi = 1.0$, $O_2/(O_2 + \mathrm{Ar})$ % vol = 5.0%, (h) H_2/O_2/Ar at $\phi = 1.0$ for fixed $U_{ign} = 160$ V. (i) n-C_4H_{10}/air at $\phi = 0.7$, (j) n-C_4H_{10}/air at $\phi = 2.2$, (k) n-C_4H_{10}/air for fixed $U_{ign} = 160$ V. Initial pressure and temperature for all tests are 1 atm and 298 K. The Le shown in is defined to be the effective Lewis number of a combustion mixture, based on theory in Ref. [24]. The spark gap distance d_g is 0.58 mm for case (a), 0.30 mm for cases (b)–(h), and 0.80 mm for cases (i)–(k). Here we identify three outcomes. Failed ignition (red crosses), where ignition kernel fails to grow to a propagating flame, either due to lack of ignition energy in quiescent environment or faster dissipation by turbulence. Successful ignition (green dots), where ignition kernel containing sufficient energy grows to a flame with or without the presence of turbulence. Ignition facilitated by turbulence (green dots with blue borders), where ignition kernel grows to a flame in the presence of turbulence, but fails to do so in quiescence if supplied with the same ignition energy.

unity. In particular, turbulence facilitates ignition with He, a light inert which substantially increases the thermal conductivity and thus Le, while with heavier inerts such as CO_2 and Ar, Le becomes less than or near unity and as a result ignition is inhibited. These extensive sets of experiments therefore rule out the possibility that the distinct turbulence effects for fuel-lean and fuel-rich conditions are due to different chemical kinetics.

Finally, in order to provide even further substantiation of the phenomena and concept advanced herein, and for fuels other than H_2, we have flipped the lean versus rich aspect of the mixture by using n-butane (n-C_4H_{10}) as the fuel, whose Le are greater and smaller than unity for lean and rich mixtures because of the substantially larger molecular weight of n-butane relative to that of oxygen. Thus lean (rich) n-butane/air mixtures should exhibit behavior similar to those of rich (lean) H_2/air mixtures, respectively.

Figures 3(i)–3(k) summarize the ignition test results for a lean and a rich n-butane/air mixture, whose Le's are 2.1 and 0.9, respectively. It is seen that ignition is indeed facilitated for the Le > 1, lean mixture. The above extensive results therefore demonstrate that ignition enhancement by turbulence for mixtures with large Le is of a general nature, irrespective of fuel, equivalence ratio, and inert.

In summary, while turbulence is usually believed to suppress ignition due to the enhanced dissipation of localized ignition energy, we have experimentally demonstrated that it can actually facilitate ignition under conditions in which ignition is limited by the difficulty of the flame kernel to transition into an expanding flame. This is possible through an embryonic flamelet structure consisting of segments subjected to both positive and negative stretch, while quiescent ignition generates only positive stretch through the positive curvature over the entire expanding, smooth flame surface. Such an understanding is of both practical and scientific significance. For example, explosion tests in quiescence may underestimate the risk of accident, while engine flows can also be optimized to reduce misfire for ultralean, clean, fuel-efficient operations, recognizing that engine fuels are large hydrocarbons such that their lean burning corresponds to Le > 1 situations. Furthermore, while studies of the supernova explosion have always assumed the initial existence of a flame that subsequently transitions to a detonation wave, perhaps it would be of interest to also investigate situations for which the establishment and sustenance of such a flame may not be possible in the first place, recognizing the fact that the Le for supernovae is exceedingly large [7], of the order of 10^4. Finally, the potential implication to the ignition and reactions of liquid-phase and supercritical systems for various technological applications, such as materials synthesis [31,32], is also of interest because they are also characterized by extremely large values of Le.

This work was supported the Air Force Office of Scientific Research.

*Corresponding author.
cklaw@princeton.edu

[1] J. C. Kantor, Science **224**, 1233 (1984).
[2] R. D. Reitz, Combust. Flame **160**, 1 (2013).
[3] F. N. Kissell, A. E. Nagel, and M. G. Zabetaki, Science **179**, 891 (1973).
[4] V. N. Gamezo, A. M. Khokhlov, E. S. Oran, A. Y. Chtchelkanova, R. O. Rosenberg, Science **299**, 77 (2003).
[5] S. E. Woosley, A. R. Kerstein, and A. J. Aspden, Astrophys. J. **734**, 37 (2011).
[6] Y. Gao and C. K. Law, Phys. Rev. Lett. **107**, 171102 (2011).
[7] S. I. Glazyrin, S. I. Blinnikov, and A. Dolgov, Mon. Not. R. Astron. Soc. **433**, 2840 (2013).
[8] D. R. Ballal and A. H. Lefebvre, Proc. R. Soc. A **357**, 163 (1977).
[9] C. F. Kaminski *et al.*, Proc. Combust. Inst. **28**, 399 (2000).
[10] D. Bradley, C. G. W. Sheppard, I. M. Suardjaja, and R. Woolley, Combust. Flame **138**, 55 (2004).
[11] S. Ahmed and E. Mastorakos, Combust. Flame **146**, 215 (2006).
[12] S. F. Ahmed, R. Balachandran, and E. Mastorakos, Proc. Combust. Inst. **31**, 1507 (2007).
[13] S. S. Shy, C. C. Liu, and W. T. Shih, Combust. Flame **157**, 341 (2010).
[14] C. Cardin, B. Renou, G. Cabot, and A. M. Boukhalfa, Combust. Flame **160**, 1414 (2013).
[15] C. K. Law, *Combustion Physics* (Cambridge University Press, Cambridge, England, 2006).
[16] P. Ronney and G. I. Sivashinsky, SIAM J. Appl. Math. **49**, 1029 (1989).
[17] J. K. Bechtold and M. Matalon, Combust. Flame **119**, 217 (1999).
[18] C. J. Sun and C. K. Law, Combust. Flame **121**, 236 (2000).
[19] Z. Chen and Y. Ju, Combust. Theory Modell. **11**, 427 (2007).
[20] P. Pelce and P. Clavin, J. Fluid Mech. **124**, 219 (1982).
[21] M. Matalon and B. J. Matkowsky, J. Fluid Mech. **124**, 239 (1982).
[22] P. Clavin and P. Garcia, J. Mech. Theor. Appl. **3**, 245 (1983).
[23] J. K. Bechtold and M. Matalon, Combust. Flame **127**, 1906 (2001).
[24] M. Matalon, C. Cui, and J. K. Bechtold, J. Fluid Mech. **487**, 179 (2003).
[25] P. Clavin and J. C. Graña-Otero. J. Fluid Mech. **686**, 187 (2011).
[26] M. L. Frankel and G. I. Sivashinsky, Combust. Sci. Technol. **40**, 257 (1984).
[27] B. Deshaies and G. Joulin, Combust. Sci. Technol. **37**, 99 (1984).
[28] L. He, Combust. Theory Modell. **4**, 159 (2000).
[29] Y. B. Zel'dovich, G. I. Barenblatt, V. B. Librovich, and G. M. Makhyiladze, *The Mathematical Theory of Combustion and Explosions* (Springer, New York, 1985).
[30] S. Chaudhuri, F. Wu, D. Zhu, and C. K. Law, Phys. Rev. Lett. **108**, 044503 (2012).
[31] S. Teragawa, K. Aso, K. Tadanaga, A. Hayashia, and M. Tatsumisago, J. Mater. Chem. A **2**, 5095 (2014).
[32] M. Singh, M. Kumar, F. Štěpánek, P. Ulbrich, P. Svoboda, E. Santava, and M. L. Singla, Adv. Mat. Lett. **2**, 409 (2011).