Non-linear response of the flame velocity to moderately large curvatures in laminar jet flames of methane–air mixtures

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ABSTRACT

An experiment was set up to investigate the correlation between flame velocity and flame stretch in moderately stretched laminar jet flames of methane–air mixtures at different equivalence ratios. Gas flow velocities were obtained by Particle Image Velocimetry (PIV) from images of axial sections of seeded flows, and the location of the flame front was related to the gradient of luminosity. Flame stretch was determined and shown to be mainly due to flame curvature, with a small contribution from the gas strain rate. Inlet gas velocities not much larger than the planar flame velocity led to short rounded flames for which the classical Markstein relation was experimentally validated at any point of the flame fronts. However, the Markstein relation ceases to be uniformly valid for larger inlet gas velocities, in the order of ten times the planar flame velocity, for which the flame adopts a slender shape. These tall flames are nearly conical and obey the Markstein relation far from their tips, but a superlinear increase of flame velocity with stretch is observed when the tip is approached. Furthermore, the local velocity of these flames depends not only on the stretch but also on the inlet velocity of the gas. This behavior has been analyzed in terms of an empirical correction to the Markstein relation that is quadratic in the flame stretch with a coefficient that depends on the gas inlet velocity. The experimental results suggest that this coefficient is zero below a certain threshold value of this velocity and increases as the square root of the excess of gas inlet velocity above this threshold. This kind of sharp transition controlled by the velocity of the incoming gas is reminiscent of a typical critical behavior and is tentatively taken to reveal a stability exchange between rounded (weakly curved) and slender (strongly curved) flames.

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1. Introduction

Laminar methane–air jet premixed flames display axisymmetric concave configurations where the increase of flame velocity by flame compression (negative stretch) leads to robust flame tips that can withstand gas inlet velocities several times larger than the planar flame velocity. This situation is thought to be common to the concave flamelets of wrinkled flames of nearly equidiffusional species distorted by turbulent flows, which gives to the laminar jet flames an exceptional interest as simple models of turbulent flames [1–4]. Preferential diffusion effects, in particular tip opening due to lack of a deficient reactant with low Lewis number at the tip of the flame, are important and have been given much attention in the literature [5–10]. The last article cited is a recent work

by Wang et al. where tip opening of syngas Bunsen flames is characterized in terms of a local Karlovitz number which represents the ratio of the characteristic reaction time of a stretched flame to the characteristic tangent flow time of this same flame. These effects, however, are beyond the scope of this paper and will not be discussed in what follows.

Jet flames have been the subject of numerous theoretical analyses, particularly after the pioneering works by Sivashinsky [11] and Buckmaster [5], who first applied activation energy asymptotics (AEA) to this problem. Asymptotic analyses have been also performed for planar flames subjected to perturbations whose characteristic length scale Λ is large compared to the planar flame thickness δ_p [12–15], using AEA to account for the reactive layer. Our aim is to use this theory to interpret the measurements obtained in methane–air jet flames. It is pertinent to emphasize here the basic assumption of this two-scale method: the inner structure of the flame is solved on the scale δ_p whereas the outer isothermal (upstream and downstream) regions are solved on the scale Λ . The

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inner solution is then used to derive jump conditions for the outer solutions through the flame front, whose location is chosen to be at the reactive sheet of the flame. These outer solutions are Taylor-expanded around the front and the results of the theory apply to these isothermal fields extrapolated to the front. To be coherent with this methodology, the experimental measurements of the gas velocity have to be performed in the isothermal zones outside of the flame and then extrapolated to the reactive layer to compare with the theoretical relations; see, e.g., [16,17]. In particular, trying to match these relations with velocity measurements done inside the thermal flame thickness (for instance, by PIV with solid tracers) is an erroneous procedure as these measurements should correspond with the theoretical solution in the inner non-isothermal preheating region.

Theoretically, for a one-step Arrhenius kinetics and to first order in terms of the ratio $\epsilon = \delta_p/\Lambda \ll 1$, the local flame velocity, U_n , was found to differ from the planar flame velocity, U_p , by relative amounts proportional to the total flame stretch [12–15]:

$$\frac{U_n}{U_p} - 1 = \frac{L}{U_p} S + O(\epsilon^2) \quad (1)$$

with

$$S = U_p \nabla \cdot \mathbf{n} + \mathbf{n} \cdot \nabla \mathbf{v}_g \cdot \mathbf{n} \quad (2)$$

for a stationary flame. Here $\mathbf{n}$ is the unit vector normal to the flame front pointing towards the burnt gas, so that $\nabla \cdot \mathbf{n}$ is twice the mean curvature of the front k_{av} , and $\mathbf{v}_g$ is the velocity field of the fresh gas upstream of the flame, so that $\mathbf{n} \cdot \nabla \mathbf{v}_g \cdot \mathbf{n}$ (evaluated at the flame front) is the rate of strain of the front due to the flow in which it is immersed. The proportionality factor L is the Markstein length, defined here in the fresh gas, which fully characterizes the flame dynamics in this limit of weak stretch ($\epsilon \ll 1$). For convenience, we call flame stretch to the magnitude S , which is the negative of the usual definition. This S is positive at any point of a jet flame. It is the factor in braces in Eq. (28) of Ref. [12] multiplied by U_p . The value of the Markstein length is determined by the theoretical analysis as a function of the physico-chemical properties of the gas mixture. The ratio of the Markstein length to the planar flame thickness is the Markstein number, $Ma = L/\delta_p$, which is the key control parameter determining the morphological stability of the planar flame [18,19]. For planar horizontal or inclined fronts propagating downwards in the gravity field, positive values of the Markstein numbers larger than some critical value, $Ma > Ma_c$, ensure smooth flame fronts that do not develop wrinkles or cells [2,18–21]. In jet flames a small nozzle favors smooth axisymmetric fronts, as it provides a cut-off for the instability wavelength that prevents the appearance of polyhedral or other non-smooth flame surfaces.

For most flammable, nearly equidiffusional gas mixtures, the Markstein length L is positive [18,20,22–24]. Thus, while positively stretched flames ($S < 0$) are retarded and eventually extinguished by nonlinear stretch effects [25], negatively stretched jet flames ($S > 0$) are concave toward the fresh gas and have velocities that increase towards the tip. According to Eq. (1), this allows jet flames to overcome gas feeding velocities larger than U_p , preventing tip opening [6,9].

We have set up an experimental facility where the local flame velocity U_n and the local mean curvature k_{av} of a jet flame have been simultaneously measured [26,27], allowing also the experimental determination of the planar flame velocity and the Markstein length. Thus, the validity of Eq. (1) has been checked at any point of the flame front, from the rounded tip to the quasi-conical skirt (Fig. 1). The aim of the present work is to carry out a systematic study on the region around the tip of moderately curved

Fig. 1. Sketch of the flame geometry: z and r are the axial and radial coordinates, respectively, U_{in} is the inlet gas velocity and $\mathbf{n}$ is the unit normal vector at any point on the flame surface. The curvature radius of the meridional section through a point is R_m whereas R_c is the curvature radius of the conical surface tangent on the same point. These are the two principal normal sections of the surface, and $\nabla \cdot \mathbf{n} = R_m^{-1} + R_c^{-1} = 2k_{av}$.

flames, where Eq. (1) is no longer valid and is expected to be replaced by some nonlinear relation.

The first reference to the limitations of the Markstein relation (1) dates back to 1960, when Petersen and Emmons [28] reported the inaccuracy of this relation for the concave intermediate tip of W-shaped methane–air flames when $(U_n/U_p - 1) \geq 0.73$. Since then much effort has been devoted to extend the velocity-stretch relation (1) and (2) in different manners, in particular to strongly stretched flames, including the possibility of extinction. Sticking to the essence of Markstein ideas, Echekki and Mungal [29] proposed an empirical modification of (2) by using the current flame velocity U_n instead of the velocity of the unstretched flame, U_p , to evaluate the contribution of the curvature to the stretch. This results in a nonlinear velocity-stretch relation that fits their measurements at the tip of a planar jet flame and predicts a maximum possible curvature of the front. On the basis of their experimental and numerical results, and of the results of a new quasi-unidimensional model, Poinso et al. [30] proposed a characterization of stretched flames in terms of two speeds; a consumption speed that is a measure of the reaction rate integrated across the flame, and a displacement speed of the flame front relative to the fresh gas. These authors postulate that the displacement speed is affected by three mechanisms; a chemical mechanism associated to changes in the structure of the reaction region, a hydrodynamic mechanism associated to area changes by lateral flow divergence and flame curvature, and a diffusive mechanism associated to misalignment of diffusion and convection fluxes. Based on their experimental and numerical results, they proposed correlations of the flame displacement speed with the flame stretch.

It has long been recognized that using a single Markstein length to characterize the contributions of the curvature and the strain rate to the flame stretch may be an oversimplification [30–32], specially in cases when a kinetic scheme more realistic than the single overall reaction assumed in the original analyses makes the location of the main reaction layer and the diffusivi-

ties of the reactants reaching this layer less straightforward than in those analyses. Bradley et al. [31] clearly described this effect by comparing numerical results obtained for stationary spherical flames and for spherical flames propagating inwards or outwards. Clavin and Graña [32] extended the analyses of [12–14], showing that two different Markstein lengths should be used to account for the two different contributions to the flame stretch, and computed these lengths for two-step chain branching mechanisms suitable for rich hydrogen-air and lean hydrocarbons-air flames. Ibarreta and Driscoll [33] and Ibarreta et al. [34] carried out experiments with inwardly propagating spherical flames whose stretch is entirely due to the curvature of the front, and found values of the Markstein length significantly larger than those of outwardly propagating flames for which the contribution of the flow strain rate to the stretch is important. Sinibaldi et al. [35] rationalized in similar terms the dispersion in the values of the Markstein length obtained for a given mixture in different experiments. An additional difficulty here is that the definition of flame velocity used by these authors does not seem to be entirely consistent with that of the analysis; see, e.g., Refs. [12,36] for a discussion of the effect of these differences on the measured Markstein length. In our experiments, the stretch is dominated by the contribution of the curvature of the front, so we may focus on a single Markstein length. Mueller et al. [37] (see also references therein) investigated the strong unsteady stretch of a flame by a vortex. They concluded that the peak of the gas dilatation rate in each section of the flame may be an indicator of the local flame strength, and included the dilatation rate in an extended definition of stretch that uses conditions inside the flame. Choi and Puri [38,39] studied separately the effect of the strain rate and the curvature in different regions of planar and axisymmetric Bunsen flames, around the tip and away from it. On the basis of their results, these authors proposed a modification of the classical velocity-stretch relation in which the flame velocity in regions of strong curvature around the tip is different from the unstretched flame velocity even in the absence of strain rate. Additional experiments and analyses of the effect of flame stretch for inward propagating cylindrical and spherical flames, and for flames in Hele-Shaw cells have been carried out in Refs. [40–42], while Halter et al. [43] proposed nonlinear velocity-stretch relations for outward propagating spherical flames.

An analysis of the structure of the tip of a very slender Bunsen flame in conditions such that the local curvature of the reaction sheet is large compared to the inverse of the thickness of the corresponding planar flame has been carried out by Buckmaster and Crowley [44] building on the previous work of Buckmaster [5]. Even though this extreme condition is not realized in our experiments, the scenario envisaged in this analysis defines a limit to the notion of a local velocity-stretch relation underlying Markstein type analyses.

2. Experimental setup and techniques

The laminar jet burner consists of a plenum chamber where fluxes of gases, coming from different feeding lines and controlled by Bronkhost High-Tech mass flow controllers, are mixed. The gas mixture is fed in a vertical tube which is fitted with several honeycomb structures and damping grids to get a uniform plug flow that exits through a convergent nozzle of 1 cm final diameter. The flame attaches to the rim of this nozzle thanks to the stabilizing effect of an annular coflow of a lean methane-air mixture, which sustains a secondary flame surrounding the primary flame of interest. By adjusting the velocity and composition of this coflow, the mixing layer around the primary flame could be controlled and flame flickering was very much reduced. The thickness of the nozzle's wall is 0.5 mm. The width of the annular jet is 1.4 cm, the equivalence ratio of the coflow stream ranges from 0.65 to 0.75,

and its velocity from about 20 cm/s to about 50 cm/s. The power released by the coflow flame is in the range of 0.5–1.5 kW, assuming methane is completely consumed. The power released by the main flame varies in a wider range. It is about 1 kW for a stoichiometric mixture with an inlet velocity $U_{in} = 3.7$ m/s. Additionally the wall of the burner was thermalized with a water jacket kept at a constant temperature.

The velocity of the fresh gas in a vertical plane through the axis of the jet was measured with PIV, using as tracers olive oil droplets carried by the gas. The oil droplets are formed by evaporation-condensation of a 100:1 oil-water emulsion injected in a seeding chamber where the emulsion is gasified by a hot plate (500 °C) and condenses shortly afterwards to form an aerosol of fine oil droplets. This chamber is inserted in a bypass of the air line to control the amount of aerosol droplets in the main flow. When these oil droplets reach the preheating region of the flame they evaporate, serving to delineate the upstream border of this region on the PIV images of the flame. Most of the preheating region is void of droplets and appears as a dark zone which, on the burnt side, is adjacent to a luminous layer due to the optical radiation emitted by the chemical reaction. No measurable changes were observed in the flame when the seeding was shut down, implying a negligible influence of the oil aerosol on flame properties.

The PIV system consists of a 532 nm wavelength Nd:YAG laser (New Wave 120XT), a sheet-forming emission optic, a CCD camera (PCO PixelFly 1392 × 1040 pixels) with automatic macro lenses (Nikon 180 mm, f/3.5 and Tamron 90 mm f/2.8), and pulse and delay generators for synchronizing all these devices. The camera is focused perpendicularly to the laser sheet on a window of about 10 × 7 mm, giving a resolution of the order of 140 pixel/mm with the Nikon macro lens. The light sheet is vertical through the axis of the burner. Pulse delays suitable for the gas velocity and size of the region of interest are chosen following standard practice for the cross-correlation method. Cross-correlation computations are done with open software [26,27].

Methane-air mixtures with equivalence ratios ranging from 0.6 to 1.5 have been investigated, with emphasis on rich flames. The PIV camera was used without any filter so that the laser light dispersed by the oil droplets into the camera is superimposed on the CCD with the light directly emitted by the flame (mostly coming from CH and C₂ emissions [45]). The exposure time of the second picture of each PIV pair is about 100 ms, enough to capture a well-defined image of the luminous reactive region of the flame together with the laser light dispersed by the oil droplets. These pictures allow us to simultaneously measure fresh gas velocity and flame shape [26,27]. Working with full aperture, the optical setup had a depth of field of about 1 mm, so most of the information in the images came from a thin slice of the flame. Therefore, the standard Abel inverse-transform was deemed unnecessary and raw images of flame slices through the tip were processed with an in-house edge detection algorithm based on a Laplacian-of-Gaussian filter. The maximum (absolute) gradient of luminosity at the rear boundary of the luminous region of the flame was extracted and identified with the theoretical AEA location of the chemical reactive sheet. This criterion allows a fast determination of the reactive sheet and represents a convenient compromise between accuracy and computational speed. When compared with the barycenter rule proposed by Davis et al. [17] in finely resolved flames with detailed chemical kinetics [47], this criterion leads to deviations of the reactive sheet location that are 20–30% of the actual reactive layer thickness. A fifth order spline was used to fit the reactive sheet position along the luminous flame front and to compute its local geometrical properties (normal vector and curvature) assuming that the flame is axisymmetric. A more sophisticated front extraction method was proposed by Guiberti et al. [46] based on

an intensified camera equipped with a bandpass filter centered at a UV emission peak of one of the radicals present in the reaction region. Using a suitable fit of the broadband CO₂ spectrum in the range of wavelengths of the filter, these authors were able to subtract the CO₂ background from the measured intensity, thereby removing the uncertainty due to the radiation of this species. We have checked that radiation from CO₂ does not invalidate our front detection algorithm based on the gradient of luminosity.

3. Numerical methods

Numerical computations have been carried out using a simple model of the flow around the tip of a tall Bunsen flame. Rather than reproducing the experimental results, the purpose of these computations is to define a minimal model that can qualitatively account for these results.

To this end, combustion is modeled by a single irreversible Arrhenius reaction that consumes the minority reactant at a rate (mass consumed per unit volume and time) $w = \rho Y B \exp(-T_a/T)$, where ρ and T are the density and temperature of the gas, Y is the mass fraction of the minority reactant, and B and T_a are the frequency factor and the activation temperature of the reaction. The reaction generates a heat q per unit mass of reactant consumed. A Fick law is assumed for the diffusion of this reactant. The viscosity (μ) and thermal conductivity (λ) of the gas, and the diffusivity of the reactant (D), increase with temperature as $\mu/\mu_u = \lambda/\lambda_u = \rho D/\rho_u D_u = (T/T_u)^\kappa$, where $\kappa = 0.75$ and the subscript u denotes conditions of the inlet fresh gas. Acoustic effects are left out using the quasi-isobaric form of the equation of state $\rho T = \rho_u T_u$, where, in addition, the mean molecular mass of the gas is taken to be constant. No body forces are considered.

Stationary axisymmetric solutions are sought of the mass, momentum, energy and minority reactant conservation equations in this zero Mach number limit. Strictly, the flow around the Bunsen flame should be computed together with the shear layer that issues from the burner rim, and the coflow, secondary flame and outer inert flow described above. This problem can be simplified for large values of the ratio U_{in}/U_p of the inlet velocity to the velocity of the planar unstretched flame, and of the fresh-to-burnt gas density ratio. Then the flame front away from the tip is close to a cone of small semiangle, and its presence induces only a small perturbation to the fresh gas flow. An asymptotic analysis [49,50] shows that the velocity of the burnt gas far from the tip is the superposition of a uniform axial flow and the flow induced by a line of sources along the axis of the flame. The pressure of the burnt gas on this region of the flame front is the sum of a uniform pressure that does not change the velocity of the fresh gas and a small variable pressure that causes a vertical acceleration of this gas and a slight deformation of the conical flame front. Only a weak vorticity is generated in this region of the flame.

This asymptotic description of the flow far from the tip can be used to set up approximate boundary conditions that allow us to compute the solution in a region around the tip of size large compared with the minimum curvature radius but small compared with the size of the whole flame. A cylindrical computational domain of radius R and height H (at the symmetry axis) is considered. The mass and momentum conservation equations are rewritten in the equivalent stream function/vorticity formulation. The flame front is artificially anchored at a circle on the surface of the cylinder, at a certain distance νR above its lower base, by locally increasing the temperature. Dirichlet conditions $T = T_u$, $Y = Y_u$, $\omega = 0$ and $\psi = \frac{1}{2} \rho_u U_{in} r^2$, where ω and ψ are the vorticity and the stream function, and r is the distance to the symmetry axis, are used at this base and at the cylindrical surface upstream of the flame. At the rest of the boundary, conditions of zero normal

derivative are used for T , Y and ω , while the stream function is set to that of the asymptotic flow described in the previous paragraph.

The adiabatic flame temperature is $T_b = T_u + qY_u/c_p$, where c_p is the specific heat of the gas, taken to be constant. The dimensionless parameter $\gamma = (T_b - T_u)/T_u$ measures the thermal expansion of the gas at the flame, and the Zeldovich number $\beta = T_b(T_b - T_u)/T_u^2$ measures the temperature sensitivity of the chemical reaction. Values of B and T_a are chosen for the well-known asymptotic expression of the planar flame velocity [15] $U_p^\infty = (2D_u B)^{1/2} \text{Le} \beta^{-1} (1 + \gamma)^{\kappa/2-1} \exp[\beta(1 + \gamma)/2\gamma]$ to fit the velocity of a planar methane-air flame in a range of equivalence ratio that extends from about $\varphi = 0.9$ to about $\varphi = 1.5$. This leads to $\beta = 62.5 \gamma/(1 + \gamma)^2$. Here $\text{Le} = \lambda_u/\rho_u D_u c_p$ is the Lewis number of the minority reactant, which is taken to be equal to unity. The problem is written in dimensionless form scaling velocities with U_p^∞ , distances with $\delta_p^\infty = \lambda_u/\rho_u U_p^\infty c_p$, ρ and Y with ρ_u and Y_u , and introducing the dimensionless temperature $\theta = (T - T_u)/(T_b - T_u)$. The conservation equations are then rewritten in the new independent variables (ξ, r) with $\xi = z/[h(r) + \nu R]$. Here z is the vertical distance above the lower base of the computational domain, ν is an adjustable constant, and the function $h(r)$, with $h(R) = 0$, is iteratively chosen to ensure that $\theta(\xi = 1, r) = 0.7$ in a central region around the flame tip. The equations are discretized using second order finite differences in a rectangular non-uniform grid that covers the domain $[0, \xi_\infty] \times [0, R]$ and is finer around $\xi = 1$, and solved by pseudo-transient iteration. (Note that the upper boundary of the computational domain, $\xi = \xi_\infty$, is no longer a planar surface.) Typical values of the numerical parameters are $R = 90$, $\nu = 1$ and $\xi_\infty = 4$. Numerical tests show that grid independence is obtained with 360×120 grid points.

4. Results

4.1. Experimental results

The velocity field of the fresh gas was determined with the PIV system in flames of various equivalence ratios and inlet velocities. As shown in [26], the main feature of this isothermal flow is a vertical acceleration of the gas which, by continuity, entails a radial flow converging towards the axis. In agreement with numerical simulations [26] and theoretical considerations [49,50], this flow is well approximated by a simple axisymmetric stream function of the form (leaving out a constant factor ρ_u) $\psi = \frac{1}{2}(U_{in} + Cz)r^2$, for which

$$v_z = \frac{1}{r} \frac{\partial \psi}{\partial r} = U_{in} + Cz \quad \text{and} \quad v_r = -\frac{1}{r} \frac{\partial \psi}{\partial z} = -\frac{C}{2} r. \quad (3)$$

The value of C depends on the inlet gas velocity and the equivalence ratio of the mixture, being typically in the range $1\text{--}20 \text{ s}^{-1}$ and decreasing with increasing flame height. The contribution of this strain rate to the flame stretch is, according to Eq. (2),

$$\mathbf{n} \cdot \nabla \mathbf{v}_g \cdot \mathbf{n} = C \left(n_z^2 - \frac{n_r^2}{2} \right), \quad (4)$$

where n_z and n_r are the axial and radial components of the normal vector $\mathbf{n}$, respectively. Close to the flame tip, $n_z \approx 1$, $n_r \approx 0$ and the contribution of the flow strain rate to the flame stretch is approximately C . Far from the tip of an elongated jet flame, $n_z \approx 0$, $n_r \approx 1$ and the contribution tends to $-C/2$. Thus, the sign of the contribution changes from positive near the tip to negative by the bottom of the flame, so that the straining effect changes from compressing (increasing flame velocity near the tip) when $n_z > n_r/\sqrt{2}$, to stretching (decreasing flame velocity near the bottom) when $n_z < n_r/\sqrt{2}$. Figure 2 shows these characteristics in the image of an actual flame tip.

Fig. 2. Left: image of a methane-air flame ($\varphi = 1.50$, $U_m/U_p = 8.5$, $L = 3$ mm) displaying the oil tracer droplets in the fresh isothermal zone and the flame symmetry axis. Right: vertical gas velocity along the symmetry axis measured by PIV. The winding curve joins the experimental measurements. The straight line is the linear regression providing the strain rate $\mathcal{C} = 2 \text{ s}^{-1}$. The dashed horizontal line marks the location of the reactive sheet at the tip.

Fig. 3. Local relation between flame velocity and twice the mean curvature $2k_{av}$ along the flame front of a methane-air flame ($\varphi = 1.12$, $U_m/U_p = 4.5$, $L = 0.7$ mm) shown in the inset. The photograph spans a region of $10 \times 7 \text{ mm}^2$ around the tip of the flame, with a resolution of $7 \mu\text{m}/\text{pixel}$. Points of the flame have been grouped in three zones denoted a, b and c. Local normal directions to the front are shown in the inset where the front position has been partially thickened to help visualization. The straight lines correspond to Markstein relation with a 95% confidence interval.

In accordance with the classical theoretical procedure explained in the Introduction, this isothermal velocity field was extrapolated through the preheating region up to the reactive sheet. A typical result, for an elongated jet flame with near-stoichiometric composition ($\varphi = 1.12$) is shown in Fig. 3. An image of the flame obtained with the 180 mm macro lens is displayed in the inset, with the detected reactive sheet marked by the outermost grey (colored) curve. This curve was used to compute the normal unit vector $\mathbf{n}$ and the mean curvature at any point. The local flame velocity was obtained at any point of this reactive sheet as the projection of the extrapolated fresh gas velocity on the normal direction $\mathbf{n}$.

This flame velocity was identified with U_n and correlated with the flame stretch at each point of the front. The strain rate effect in Eq. (2) is very small compared with the curvature effect and can be neglected, so that $S \approx 2U_p k_{av}$, with $2k_{av} = k_m + k_c$. In the figure, the error bars account for the whole chain of measurement (experimental and computational treatment of the data). The largest error comes from the determination of the curvature, which requires the computation of a second derivative. Anyway, the statistical dispersion of the results obtained in different runs under the same experimental conditions turns out to be significantly larger than these errors.

The two curves in Fig. 3 show results for the two sides of the flame axial section. Their difference is a measure of the small asymmetry of the flame cross-section. Continuously varying gray level (color) identifies location along the flame front as keyed by the gray level (color) in the inset. Three distinguished zones are displayed: (a) linear region where Markstein's relation holds, (b) near-tip zone where the relation is no longer linear, and (c) the tip region where the two curvatures collapse to $k_{av|tip} \approx 1.5 \text{ mm}^{-1}$ leading to a perceptible spherical cap.

The bottom of the flame near the nozzle is a region of weak stretch as the flame curvature is on the order of the inverse of the nozzle radius $R_{nozzle} = 5 \text{ mm}$. Images of flames obtained with the 90 mm f/2.8 macro lens were used to characterize this bottom region. The fit of the experimental results by the linear relation (1) is also shown in Fig. 3. The intercept and the slope of this straight line give the planar flame propagation velocity and the Markstein length, respectively. Flames with four different equivalence ratios, $\varphi = 0.60, 0.90, 1.12$ and 1.50 , have been characterized. In all cases, the measurements of the corresponding planar flame propagation velocities and Markstein lengths led to values in reasonable agreement with those found in the literature [16,48–50].

It is worth noticing that the value of the Markstein length was found to be close to the corresponding radius of curvature at the tip for sufficiently tall flames, $L \sim R_{tip}$. This result points towards the Markstein length as the natural scale of the distortions of (smooth) curved flames, in a similar way as the diffusive flame thickness serves to characterize planar flames. Figure 4

Fig. 4. Excess flame velocity versus dimensionless stretch along the flame front of a very rich methane–air flame ($\phi = 1.50$, $L = 3$ mm). Data of flames obtained with three different inlet velocities $U_{in}/U_p = 4, 9$ and 20 are shown. Asterisks correspond to the tips of the measured flames (more than 120 samples). These tip data are fitted with a power law (see Section 5.1) shown as a solid line labeled with stars. The Markstein relation is the straight line in the lower part of the figure.

shows the excess flame velocity over the unstretched flame velocity as a function of the dimensionless stretch, SL/U_p , for a very rich flame ($\phi = 1.50$) and several inlet velocities. The Markstein relation is a straight line of unit slope in this figure. Over 120 flames were experimentally characterized for different values of the inlet gas velocity. Only three typical samples are shown in the figure, representing flames with small, intermediate and large heights. The short flame was obtained for $U_{in}/U_p = 4$. The validity of the Markstein relation is confirmed everywhere on the flame front, from the bottom to the tip, with the dimensionless stretch varying in the approximate range $1 \leq SL/U_p \leq 3.5$. The flame of medium height is for $U_{in}/U_p = 9$, with the stretch varying in the range $1 \leq SL/U_p \leq 4$. The Markstein relation is still retrieved in the lower part of this flame, where $SL/U_p \leq 2$, but the local flame velocity becomes larger than predicted by the Markstein relation when the tip is approached, for $SL/U_p \geq 2$, reaching up to more than nine times the planar flame velocity at the tip. The tall flame, for $U_{in}/U_p = 20$, behaves in a similar way, but with a larger flame velocity at the tip, higher than twenty times the planar flame velocity. Note that in the last two cases, when the velocity-stretch relation ceases to be linear, the local flame velocity is not determined by the local stretch only but depends also on the inlet gas velocity. For a given value of the stretch, the flame velocity increases with the inlet velocity. For instance, the flame with $U_{in}/U_p = 9$ shows a flame velocity $U_n/U_p \approx 5$ at the place where the stretch is $SL/U_p = 3$, whereas, under the same stretch, the flame with $U_{in}/U_p = 20$ shows a larger velocity $U_n/U_p \approx 6$. This implies that there is no longer a simple univocal velocity-stretch relation. Instead, the velocity with which the gas reaches the near-tip zone seems to play a role in determining the value of the flame velocity. Since the velocity of the fresh gas does not change much from the nozzle to the tip of the flame, the discussion here and in the rest of the paper is phrased in terms of the velocity U_{in} .

Small deviations of the flames from axisymmetry and steadiness (flickering) lead to significant loss of accuracy in the determination of geometrical parameters at the flame tip. The effect is apparent in Fig. 4 by the dispersion of asterisk symbols, mainly for tall flames, due to the variable behavior obtained near the flame tips. Depending on the flame sample, the observed behavior can depart from a shape like in Fig. 3, where the velocity-stretch relation shows a monotonously rising curve, to exhibit uneven curly

behaviors like the reversed C-curves of Fig. 4. Since the region affected by these unsteady effects is very small, the localized unsteadiness is not expected to have an important effect on the results.

The full expression of the flame stretch Eq. (2) has been used to draw Figs. 3 and 4. However, due to the smallness of $\mathcal{C}$, the contribution of the strain rate to the stretch, given by Eq. (4), is small compared to the contribution of the curvature. Even in short flames with small curvatures, which comply with Markstein relation, the strain rate accounts for less than 10% of the stretch at the flame tip for non-stoichiometric (slow) flames, and this contribution decreases to about 2% for near-stoichiometric (fast) flames. In both cases, the contribution is reduced further for tall flames with high curvatures, becoming 1% and 0.2%, respectively.

4.2. Numerical results

Some numerical results are shown in Fig. 5, which is to be compared with the experimental results for slender flames in Fig. 4. In the numerical computations, the reaction front is defined as the locus of point of maximum reaction rate in each section of the flame. Once this front and its local normal have been computed, the projection of the fresh gas velocity along the normal can be evaluated as a function of the distance to the front. The normal flame velocity U_n is the extrapolation of this normal gas velocity to the reaction front. Only the velocity of the cold gas, with $\theta < 0.1$, is used for this extrapolation. The flame stretch S is computed using Eq. (2). The main contribution to the stretch is due to the curvature of the reaction front.

Each curve in Fig. 5 shows the normal flame velocity as a function of the stretch for a different flame, the rightmost point of each curve corresponding to the flame tip. Results are given for two values of the thermal expansion parameter $\gamma = (T_b - T_u)/T_u$ and five values of the inlet gas velocity scaled with the planar flame velocity.

Like in the experimental results of Fig. 4, two distinct regions are apparent for each flame in Fig. 5. Away from the tip, where the curvature of the front is small, the velocity-stretch relation is nearly linear (Markstein), independently of the inlet velocity. Around the tip, where the curvature increases, this relation departs from linearity and shows the same behavior as the experimental results, whereby the inlet gas velocity affects the flame velocity. A higher thermal expansion extends the linear behavior into higher local flame velocities.

5. Discussion

5.1. Analysis of the results

The main outcome of the present work, concerning the non-linear correlation of local flame velocity with local flame stretch and its dependence on the inlet gas velocity, is obtained both from experiments and numerical computations. Despite the magnitude of the experimental errors and the simplicity of the numerical model, comparison of Figs. 4 and 5 shows good qualitative agreement between both approaches. The discrepancies should be mainly ascribed to the over-simplified chemical rate model, the finite size of the computational domain and the slightly different selection of the flame front location (the maximum chemiluminescence usually occurs at a distance of about 0.1–0.3 times the reaction zone thickness downstream of the maximum heat release rate).

Figure 6 shows the difference between the shape of a flame observed in experiments and the shape predicted by the linear Markstein relation, Eq. (1), in the same conditions. The thick curve displays the actual flame front detected experimentally, with the

Fig. 5. Excess flame velocity versus dimensionless stretch along the jet flame front. Results are shown for two values of the thermal expansion, $\gamma = (T_b - T_u)/T_u = 4.94$ (solid curves) and 9.84 (dashed curves), and the five values $U_{in}/U_p = 1.33, 2, 3, 4$ and 5, increasing as indicated by the arrow. The darker dashed line shows the Markstein relation. The upper inset is a close-up of the small stretch region, illustrating the higher accuracy of the Markstein relation for flames with larger thermal expansion. The inset on the right-hand side shows the excess flame velocity as a function of the dimensionless curvature (which is the main contribution to the stretch) for $U_{in}/U_p = 5, 10$ and 20, increasing as indicated by the arrows, and the same values of the thermal expansion. Note that, in terms of the dimensionless curvature $\delta_p \nabla \cdot \mathbf{n}$, the slope of the Markstein linear relation is the Markstein number L/δ_p , which depends on the mixture composition and thus on the thermal expansion.

Fig. 6. Comparison between the actual flame shape and the shape predicted by the linear Markstein relation for the same planar flame velocity and gas inlet conditions ($\varphi = 1.50$, $U_{in}/U_p = 8.8$, $L = 3$ mm). The Markstein length has been used as unit of length.

gray levels (colors) indicating the main parts of the flame as in Fig. 3. The thin curve is the flame front computed from Eq. (1); see [27] for details. The significant difference between the two shapes shows the increase on the flame velocity over the prediction of Markstein relation, which allows the flame tip to overcome the large gas velocity with a moderate front curvature.

A large number of flames with different inlet gas velocities ($U_{in}/U_p = 3$ –20) have been characterized for the equivalence

Table 1

Summary of experimental data for different compositions identifying the function f_{in} by the regression of Eq. (7) and the values of the critical stretch and the critical flame velocity.

φ	$LS_c/U_p = 2L/R_c$	V_c	a
0.60 ± 0.04	1.9 ± 0.2	2.8 ± 0.9	0.21 ± 0.03
0.90 ± 0.07	0.9 ± 0.4	2.2 ± 0.2	0.40 ± 0.04
1.12 ± 0.07	0.8 ± 0.4	2.0 ± 0.7	0.40 ± 0.10
1.50 ± 0.08	3.1 ± 0.5	3.3 ± 0.3	0.12 ± 0.01

ratios shown in Table 1 ($\varphi = 0.60, 0.90, 1.12, 1.50$). Figure 7 displays the results for $\varphi = 1.50$ in a log-log diagram between the local values of the relative excess of flame velocity $U_n/U_p - 1$ and the dimensionless mean curvature radius R_{av}/δ_p or R_{av}/L .

Note that the maximum dimensionless curvature radius at the tip takes values of order unity when it is normalized by the Markstein length, ($R_{av}/L = 2U_p/SL$), as shown in the horizontal upper axis. The Markstein length L is thus the natural scale to analyze the flame morphology in non-planar configurations instead of the planar flame thickness δ_p . The (blue) dots at the lower right of Fig. 7 represent the apex of different flames that follow Markstein relation whereas (blue) stars at the upper left represent nonlinear flame tips. The abrupt transition between these two data sets is made evident by the log-log plot. The tips of rounded flames follow Markstein relation (a straight line with slope equal to -1 in Fig. 7), while the tips of elongated flames follow a different trend that can be approximately fitted by a power law with an exponent close to -3 . The transition between the two behaviors takes place rather abruptly for a value of the mean curvature radius at the tip that is about eight times the planar flame thickness, $R_c \approx 8\delta_p$, or equivalently of the order of the Markstein length, $R_c \approx 0.6L$. The results obtained for flames with other equivalence

Fig. 7. Log-log representation of the relative excess flame velocity versus dimensionless mean curvature radius for a very rich methane-air flame ($\varphi = 1.50$, $L = 3$ mm) and several inlet velocities $3 < U_{in}/U_p < 20$. For the lower inlet velocities, the points (blue dots) corresponding to the flame tips are aligned with the Markstein relation. However, data for flame tips at higher inlet velocities approach a fractional power law (blue stars) as in Fig. 4. Curves of the same color represent flame fronts with the same inlet velocity, from low (red) to high (blue)

ratios show the same qualitative features although the numerical values of the exponent α introduced in Fig. 4 are slightly different. Thus $\alpha = 2.7 \pm 0.3$, 2.0 ± 0.2 , 2.2 ± 0.2 and 3.0 ± 0.3 for $\varphi = 0.60$, 0.90 , 1.12 and 1.50 , respectively.

5.2. Empirical extension of the AEA linear prediction

The asymptotic analysis leading to Eq. (1) relies on the assumption that the characteristic length of spatial variations along the flame front is large compared to the flame thickness. In the present experiments, the zone of largest front curvature around the tip has a characteristic size R_{tip} of the order of the Markstein length, and was found to be in the range from 24 (for short rich flames) to 6 (for tall lean flames) times the thermal flame thickness. Thus, the ratio of the flame thickness to the characteristic curvature radius of the flame tip (δ_p/R_{tip}) is between 0.04 and 0.17. The smallness of these values suggests that the asymptotic analysis [12–14] is applicable to our flames.

On the other hand, it is evident that Eq. (1) cannot give a satisfactory account of all the experimental observations because it does not reproduce the departure from the linear behavior observed experimentally, nor the dependence of the flame velocity on variables additional to the stretch. In principle, these non-linear effects could be explained if higher order terms of the asymptotic expansion for small ϵ introduced in [12–14] were computed to account for additional contributions of the dimensionless flame stretch to the velocity. In practice, this analysis would be extremely cumbersome and the approach seems almost hopeless within the same framework.

Here a simple empirical extension of Markstein linear relation inspired by the central idea of the theoretical expansion is proposed to try to explain the experimental results. Introducing the

Markstein number $Ma = L/\delta_p$, the local relation Eq. (1) can be rewritten as

$$V = 1 + Mas + O(\epsilon^2) \quad (5)$$

where $V = U_n/U_p$ and $s = S\delta_p/U_p$ are the dimensionless forms of the flame velocity and the stretch, respectively. The second one is of the order of the small expansion parameter, $s = O(\epsilon)$, being proportional to the ratio of the flame thickness δ_p to the characteristic size of the curved flame ($R_{av} \approx 2U_p/S$). In line with the previous discussion, it seems natural to expect that the next term of order ϵ^2 should be proportional to the square of the dimensionless stretch, $s^2 = O(\epsilon^2)$, and that the order unity coefficient multiplying s^2 should contain essentially the dependence of V on the inlet gas velocity. Thus we propose to extend the validity of Eq. (1), assuming the following form of the local velocity-stretch relation

$$V = 1 + Mas + f_{in}(Mas)^2 + O(\epsilon^3). \quad (6)$$

The new coefficient f_{in} affecting the quadratic term should be chosen to account for the experimental dependence of the flame velocity for a given stretch on the incoming velocity of the fresh gas.

If the vertical acceleration of the incoming fresh gas given by Eq. (3) were taken into account, this velocity and thus the coefficient f_{in} should be different for different regions of the flame front located at different heights. However, the experimental results show the relative smallness of the vertical velocity gradient (a few percent in Fig. 2), much smaller than the experimental noise. As mentioned above, this allows us to use the same value for the vertical fresh gas velocity at any point of the flame front. With this simplification, which is specific to our experiment, the inlet gas velocity U_{in} is the only control parameter once the equivalence ratio is given, and f_{in} should be recast as function of it; of the form $f_{in}(V_{in})$ say, with $V_{in} = U_{in}/U_p$. This function is obtained empirically by least square regression of the experimental data for the velocity-stretch local relation, measured at each inlet velocity for a given equivalence ratio. The result of this analysis for $\varphi = 1.50$ is shown in Fig. 8 where it appears that a square-root like dependence provides a good fit. In this representation, the function f_{in} is tentatively taken to be

$$f_{in} = \begin{cases} a\sqrt{V_{in} - V_c} & \text{if } V_{in} > V_c \\ 0 & \text{if } V_{in} \leq V_c \end{cases} \quad (7)$$

where V_c and a are constants for a given equivalence ratio. The parameter V_c is a threshold value of the dimensionless inlet velocity below which the nonlinear correction term in (6) vanishes and the linear Markstein relation remains valid everywhere on the flame front. When the dimensionless gas inlet velocity attains the value V_c , the flame curvature at the tip reaches the maximum value that can be correctly predicted by the Markstein relation, and corresponds to a threshold value of the stretch equal to $S_c = (V_c - 1)U_p/L$. When the value V_c is exceeded, the quadratic term increases the values of the flame velocity over the Markstein prediction at any point of the flame, depending on the local stretch.

The critical value of the parameter V_c depends on the equivalence ratio as shown in Table 1. For near-stoichiometric flames, a minimum value around $V_c \approx 2$ is found, whereas for non-stoichiometric flames, the critical value reaches $V_c \approx 3$. On the other hand, the value of the coefficient a depends slightly on the equivalence ratio, varying from 0.12 in rich flames ($\varphi = 1.5$) to 0.21 in lean flames ($\varphi = 0.6$).

Thus, according to this description, the relation between flame velocity and stretch for compressed flames with $s > 0$ changes abruptly from linear to nonlinear when the dimensionless inlet gas velocity becomes larger than the critical value V_c . This sudden variation, which in our experiment is triggered by increasing the inlet velocity, is reminiscent of a critical stability-exchange transition

Fig. 8. Values of the quadratic coefficient in Eq. (6) computed by applying this equation to the experimental results in Fig. 7 for tall flames with $\varphi = 1.5$. Here $V_{in} = U_{in}/U_p$. The solid curve is a square root fit with a 95% confidence interval.

from rounded jet flames to slender jet flames. Even though a sound theoretical basis for this kind of behavior is lacking, it is worth recalling here the analysis of Buckmaster [5] and Buckmaster and Crowley [44] for very slender jet flames, for which axial diffusion is negligible. The flame structure described by these authors corresponds to flame fronts curved on the scale of the thermal flame thickness, which are therefore far from the main hypothesis underlying the local relation Eq. (1). However, these solutions are candidates to describe the flame tips observed far in the non-linear regime, for supercritical values of the inlet velocity $V_{in} \gg V_c$, where the notion of a local velocity-stretch relation breaks down.

6. Conclusions

Experiments with axisymmetric jet flames of methane–air mixtures have been conducted to extend the relation between negative flame stretch and flame velocity beyond the range of the Markstein linear prediction. A simple model of the flame has been used to perform numerical computations. The results of both studies qualitatively agree and reveal the prevalence of the linear Markstein relation up to values of the excess flame velocity much higher than expected according to the AEA analysis. Thus, the relative flame velocity U_n/U_p can reach values around 2 or 3 in flames whose shapes are still correctly predicted by the Markstein relation. However, once the inlet gas velocity surpasses a certain critical value that depends on the equivalence ratio, $U_{in}/U_p > V_c$, the increase of the flame velocity with the stretch becomes faster than linear and values as high as $U_n/U_p \approx 20$ have been measured at the tip of tall flames, a fact already reported by Choi and Puri [39]. Furthermore, in these conditions, the flame velocity ceases to depend only on the local stretch. The actual value of the inlet gas velocity need also be supplied together with the stretch to determine the value of the flame velocity without ambiguity.

An interesting result of this work, with implications for turbulent flames, is that the minimum characteristic size of the flame tip is given by the Markstein length. This result is consistent with

the analysis of the morphological stability of a planar flame that leads to a critical wavelength which turns out to be also on the order of the Markstein length. Both cases suggest that, in locally smooth flames, the modifications of the inner flame structure by the curvature produce a low-pass filter-like effect leading to a cut-off that suppresses flame corrugations with scales smaller than the Markstein length. Denet and Joulin [51] arrived to the same conclusion from numerical experiments with an extended Michelson–Sivashinsky equation accounting for stretch. Thus this length should be considered as the typical length characterizing the smallest distortions of a flame front, for instance in the corrugated flamelet regimes in the Borghi diagram [52].

Within the experimental error, our results can be summarized in the expression

$$V - 1 = Mas + \Re(a\sqrt{V_{in} - V_c})(Mas)^2 + O(\epsilon^3). \quad (8)$$

The sharp variation from linear to nonlinear behavior that appears to be well described by a square root law may indicate the existence of an instability threshold which separates flames with rounded tips from slender flames with pointed tips, where the inlet gas velocity plays the role of the control parameter. This conclusion points towards the interest of pursuing the asymptotic analysis of [12] beyond the first order to settle the validity of the empirical extension of Markstein relation proposed here as Eq. (8). Extension of this work to other hydrocarbons with non-unity Lewis numbers is currently under way.

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Supplementary material

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.combustflame.2019.03.035.

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