Pulsating instabilities of combustion waves in a chain-branching reaction model

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Abstract

In this paper we investigate the properties and linear stability of travelling premixed combustion waves and the formation of pulsating combustion waves in a model with two-step chain-branching reaction mechanism. These calculations are undertaken in the adiabatic limit, in one spatial dimension and for the case of arbitrary Lewis numbers for fuel and radicals. It is shown that the Lewis number for fuel has a significant effect on the properties and stability of premixed flames, whereas varying the Lewis number for the radicals has only qualitative (but not qualitative) effect on the combustion waves. We demonstrate that when the Lewis number for fuel is less than unity, the flame speed is unique and is a monotonically decreasing function of the dimensionless activation energy. Moreover, in this case, the combustion wave is stable and exhibits extinction for finite values of activation energy as the flame speed decreases to zero. However, for the fuel Lewis number greater than unity, the flame speed is a C-shaped and double valued function. The linear stability of the travelling wave solution was determined using the Evans function method. The slow solution branch is shown to be unstable whereas the fast solution branch is stable or exhibits the onset of pulsating instabilities via a Hopf bifurcation. The critical parameter values for the Hopf bifurcation and extinction are found and the detailed map for the onset of pulsating instabilities is determined. We show that a Bogdanov-Takens bifurcation is responsible for both the change in the behaviour of the travelling wave solution near the point of extinction from unique to double valued type as well as for the onset of pulsating instabilities. We investigate the properties of the Hopf bifurcation and the emerging pulsating combustion wave solutions. It is demonstrated that the Hopf bifurcation observed in our present study is of supercritical type. We show that the pulsating combustion wave propagates with the average speed smaller than the speed of the travelling combustion wave and at certain parameter values the pulsating wave exhibits a period doubling bifurcation.

Suggested running head: Pulsating instabilities

1 Introduction

Premixed combustion waves in models with chain-branching kinetic mechanism have drawn the interest of researchers for a long period of time [Chao & Law, 1994; Dold, 2007; Gubernov et al., 2006a; Gubernov et al., 2008a; Gubernov et al., 2008b; Joulin et al., 1985; Liñán, 1971; Mikolaitis, 1986]. These types of models can describe propagation of combustion waves in hydrocarbon-air mixtures, which is an issue of practical importance. Hydrocarbon flames normally produce a pool of radicals through branching reaction steps. These radicals later recombine to generate heat and products. The chain-branching reaction mechanism cannot be described by an overall single gross reaction. The simplest models describing flames with chain-branching kinetics should include at least two reaction steps.

The first two-step chain branching reaction model was introduced by Zeldovich [1948] and was later analyzed by Liñán [1971] using the activation energy asymptotic (AEA) method. This model is usually referred to as the Zeldovich-Liñán model. The model comprises a chain branching reaction $A + B \rightarrow 2B$, and chain-breaking (or recombination) reaction $B + B + M \rightarrow 2P + M$, where A is the fuel, B is the radicals, P is the product, and M is a third body. It is assumed that the first reaction has large activation energy and is isothermic, whereas the recombination reaction has negligible activation energy and is exothermic. The isothermic first reaction condition was subsequently dropped in Joulin et al. [1985].

In Liñán [1971] it was shown that there are three flame regimes in the Zeldovich-Liñán model: fast, intermediate and slow recombination regimes. Depending upon the particular flame regime various asymptotic expansions have been introduced in different flame zones. The resulting asymptotic differential Eqs. are then solved either analytically or numerically depending on the complexity of the set of Eqs. obtained as a result of the asymptotic analysis. The model considered in Liñán [1971] and Joulin et al. [1985] does not include heat loss and the response curves obtained in these papers are single valued functions. In Chao & Law [1994] the Zeldovich-Liñán model with heat loss to the surroundings was considered using the AEA method. It was demonstrated that the flame speed as a function of other parameters of the problem is a C-shaped function which exhibits turning point type extinction condition similar to that predicted by the one-step nonadiabatic model [Joulin & Clavin, 1979].

In a number of papers [Seshardi & Peters, 1983; Mikolaitis, 1986; Tam, 1988b; Tam, 1988a], the influence of stretch on premixed flame for the Zeldovich-Liñán model was studied. In these papers the authors considered several distinguished limits in order to examine the problem in terms of AEA either analytically or semi-analytically. As a result it was found that the flame response to stretching depended upon the particular flame regime i.e. fast, intermediate or slow recombination.

The stability analysis of flames in the Zeldovich-Liñán model has to date not been performed. It appears that the stability of the combustion waves cannot be treated effectively by using the AEA method for this model. The complexity of the stability analysis increases significantly as the number of reactions involved in the kinetic scheme increases.

The other important property of the Zeldovich-Liñán model is the nonlinear dependence of the reaction terms on the concentration of reactants. In order to overcome this difficulty, a simplified version of the Zeldovich-Liñán model was introduced recently by Dold [2007]. In this model the order of the recombination reaction was reduced by one so that the resulting kinetic scheme is written as $A+B \rightarrow 2B$, $B+M \rightarrow P+M$. The reduction of the order of the chain-breaking reaction makes the dependence of the reaction terms on the concentration of radicals in the governing Eqs. linear, which allows the problem to be treated by using the AEA method. The speed of the combustion wave was determined as a function of the parameters of the problem and was shown to be C-shaped in the nonadiabatic case. For the adiabatic case the expression derived in Dold [2007] suggests a unique flame speed. For the case of the reactant Lewis number less than one, the analysis in Dold [2007] predicts that the wave can lose stability due to the emergence of cellular instabilities.

In our earlier paper [Gubernov et al., 2006a] we have investigated the properties of the model introduced in Dold [2007] in the adiabatic case and in the limit of equal diffusivity of the reactant, the radical and heat. In contrast to Dold [2007] the activation energy in Gubernov et al. [2006a] is taken to be $O(1)$ (not an infinite number). As is noted in Mikolaitis [1986] this is a reasonable assumption for real flames like the hydrogen oxidation flame. We also used a different nondimensionalization, which enabled us to make more convenient comparisons between the two- and one-step models. In Gubernov et al. [2006a] the properties of the travelling wave solutions were investigated in detail by means of numerical simulation. It was demonstrated that the speed of a combustion wave as a function of parameters is single valued. We have found that for finite activation energy there is a residual amount of reactant left behind the travelling combustion wave which is not used in the reaction [Gubernov et al., 2004]. This makes the problem similar to the nonadiabatic one-step premixed flames. The other characteristic of the model considered in Gubernov et al. [2006a] which makes the similarity between the adiabatic two-step reaction and the nonadiabatic one-step system even stronger is that for certain parameter values the combustion wave exhibits extinction. However, for the former case the extinction occurs at zero flame speed. This mainly distinguishes the one- and two-step adiabatic models. The route to extinction in this model is investigated in detail in Gubernov et al. [2008a]. It is shown that the flame speed as a function of activation energy approaches zero in a linear fashion. The stability of the travelling combustion waves is also investigated. We have shown that for the equidiffusional case the flame is stable for a wide parameter range considered in the paper which correlates with the results in Dold [2007].

The equidiffusional approximation used in Gubernov et al. [2006a] and Gubernov et al. [2008a] makes the analysis of the problem more convenient. However, this distinguished limit reduces the applicability of the results to real flames with chain-branching reaction mechanism, which can be characterized by various values of Lewis numbers for both radicals, L_B, and fuel, L_A. This is especially true for the stability analysis, since the flame stability is expected to depend substantially on these parameters [Dold, 2007]. In our recent paper [Gubernov et al., 2008b] we investigate the effect of the Lewis number variation on both properties and the stability of combustion waves in this model. It is shown that the Lewis number for fuel has a significant effect on the properties and stability of premixed flames, whereas variation of the Lewis number for the radicals has only quantitative (but not qualitative) effect on the combustion waves. We demonstrate that, when the Lewis number for fuel is less than unity, the flame speed is a unique, monotonically decreasing function of the dimensionless activation energy. The combustion wave is stable and exhibits extinction for finite values of activation energy as the flame speed decreases to zero. For the fuel Lewis number greater than unity the flame speed is a C-shaped and double-valued function. The slow solution branch is shown to be unstable whereas the fast solution branch is stable or exhibits the onset of pulsating instabilities via the Hopf bifurcation.

In Gubernov et al. [2008b] only several characteristic values of L_A and L_B were considered which limits the understanding of the stability of the combustion waves. The aim here is to obtain a detailed map of the onset of pulsating instabilities in this reaction scheme. This includes the analysis of the various bifurcations and scenarios leading to instabilities. In particular we study the properties of the Hopf bifurcation leading to the onset of pulsations in detail and investigate the pulsating solutions emerging as a result of this bifurcation. We also investigate the existence of the Bogdanov-Takens bifurcation which is shown to play a significant role in the emergence of oscillations in both premixed [Gubernov et al., 2004] and diffusion flames [Gubernov & Kim, 2006] with single-step kinetics. The paper is organized as follows. In the next Sec. the governing Eqs. and the boundary conditions are introduced. In Sec. 3 the properties of the travelling combustion waves are investigated in detail. Section 4 is devoted to the linear stability analysis of the travelling combustion waves. The pulsating combustion wave solutions and the properties of the Hopf bifurcation are studied in Sec. 5. In the final Sec. a summary of the results and concluding remarks are presented.

2 Model

We consider an adiabatic model for premixed flame propagating in one spatial dimension that includes two steps: autocatalytic chain branching $A + B \rightarrow 2B$ and recombination $B + M \rightarrow C + M$. Here A is the fuel, B is radicals, C is the product, and M is a third body. It is assumed that all the heat of the reaction is released during the recombination stage and the chain branching stage does not produce or consume any heat. Following Gubernov et al. [2008a], the governing Eqs. for the nondimensional temperature, u, concentration of fuel, v, and radicals, w, can be written in nondimensional form as

$$\begin{aligned} u_t &= u_{xx} + rw, \\ v_t &= L_A^{-1} v_{xx} - \beta v w e^{-1/u}, \\ w_t &= L_B^{-1} w_{xx} + \beta v w e^{-1/u} - r\beta w, \end{aligned} \tag{1}$$

where x and t are the dimensionless spatial coordinate and time respectively, L_A and L_B are the Lewis numbers for fuel and radicals respectively, β is the dimensionless activation energy of the chain-branching step (which corresponds to the definition for the one-step model [Gubernov et al., 2004]), r is the ratio of the characteristic time of the recombination and branching steps (which cannot be reproduced in one-step approximations of the flame kinetics).

Equations (1) are considered subject to the boundary conditions

$$\begin{aligned} &u = 0, \qquad v = 1, \qquad w = 0 \qquad \text{for} \qquad x \rightarrow \infty, \\ &u_x = 0, \qquad v_x = 0, \qquad w = 0 \qquad \text{for} \qquad x \rightarrow -\infty. \end{aligned} \tag{2}$$

On the right boundary we have cold ($u = 0$) and unburned state ($v = 1$), where the fuel has not been consumed yet and no radicals have been produced ($w = 0$). The nondimensionalized ambient temperature is taken to be equal to zero. On the left boundary ($x \rightarrow -\infty$) neither the temperature of the mixture nor the concentration of fuel can be specified. We only require that there is no reaction occurring so the solution reaches a steady state of (1). Therefore the derivatives of u, v are set to zero and $w = 0$ for $x \rightarrow -\infty$.

The solution to the problem (1) and (2) is sought in the form of a travelling wave $u(x, t) = u(\xi)$, $v(x, t) = v(\xi)$, and $w(x, t) = w(\xi)$, where a coordinate in the moving frame, $\xi = x - ct$, is introduced and c is the speed of the travelling wave. Substituting the solution of this form into the governing Eqs. we obtain

$$\begin{aligned} &u_{\xi\xi} + cu_\xi + rw = 0, \\ &L_A^{-1} v_{\xi\xi} + cv_\xi - \beta v w e^{-1/u} = 0, \\ &L_B^{-1} w_{\xi\xi} + cw_\xi + \beta v w e^{-1/u} - r\beta w = 0. \end{aligned} \tag{3}$$

The boundary conditions (2) can be modified if we multiply the first Eq. in (3) by β, add it to the second and third Eqs. in (3) and integrate it once over ξ from minus to plus infinity. This yields a condition: $\lim_{\xi \rightarrow -\infty} S = \lim_{\xi \rightarrow +\infty} S$, where $S = \beta u + v + w$. Combining this condition with (2) results in

$$\begin{aligned} &u = 0, \qquad\qquad\qquad v = 1, \qquad w = 0 \qquad \text{for} \qquad \xi \rightarrow \infty, \\ &u = \beta^{-1}(1 - \sigma), \qquad v = \sigma, \qquad w = 0 \qquad \text{for} \qquad \xi \rightarrow -\infty, \end{aligned} \tag{4}$$

where σ denotes the residual amount of fuel left behind the wave and is unknown until a solution is obtained.

As shown in [Gubernov et al., 2008a] in order for the travelling combustion wave solution to (1) to exist the following condition has to be satisfied

$$r > \sigma \exp\left(\frac{-\beta}{1-\sigma}\right). \tag{5}$$

The inequity (5) implies that in the product region the reaction is completed and the branching term is less than the recombination term in the third Eq. of (1). The condition (5) defines the region in the parameter space where the autowaves exist.

3 Properties of the Travelling Combustion waves

The properties of the travelling combustion waves were investigated numerically by solving the system of Eqs. (3) subject to boundary conditions (4). We used a standard shooting algorithm with a fourth order Runge-Kutta integration scheme in the first instance and then the results were corrected by employing the relaxation algorithm. The investigation of the travelling combustion waves in Gubernov et al. [2008b] by using the methods described above revealed that the Lewis number for fuel L_A has a substantial effect on the properties of the premixed flames whereas the variation of L_B effects only quantitative behavior of the combustion waves. The results obtained in Gubernov et al. [2008b] and in the course of our current work are summarized in Fig. 1 where the flame speed is plotted as a function of β for various values of L_A and L_B as shown in the Fig. captions and for $r = 0.001$. Throughout this paper parameter r is fixed at this value (unless stated otherwise).

For the case of Lewis number for fuel less than unity ($L_A < 1$) the dependence $c(\beta)$ is a monotonically decaying function exhibiting extinction as the flame speed reaches zero at a certain value of the activation energy, β_e, corresponding to extinction. The flame speed decreases to zero according to a quadratic law i.e. $c \sim (\beta - \beta_e)^2$ as the dimensionless activation energy approaches the extinction value β_e. In Fig. 1 the dependence of $c(\beta)$ is plotted for $L_A = 0.1$ and $L_B = 1.0$ and 10.0. The individual curves plotted for $L_B = 1.0$, and 10.0 possess the same qualitative behaviour although the values of the flame speed change with variation in L_B. The dependence of $c(\beta)$ is a monotonically decaying function exhibiting extinction for β around 3.2. We have also found (not shown here) that the dependence of σ on β is a single valued function for $L_A < 1$. For parameter values sufficiently away from the extinction condition the residual amount of fuel can be neglected. Almost all fuel is converted to radicals and no fuel leakage is observed ($\sigma \to 0$). As we increase β and approach the extinction point, the value of σ becomes significant. At the extinction condition the residual amount of fuel reaches its maximum value corresponding to the extinction condition defined by (5).

For the case $L_A = 1$ the structure of the travelling solution branch in the parameter space changes. In Fig. 1 the dependence of the flame speed, c, on β is plotted for $L_B = 1.0$, and 10.0. Although $c(\beta)$ is still a monotonic function approaching zero as β reaches some critical value $\beta_e = 4.2...$ corresponding to extinction, the behaviour of this function is now different near the point of extinction. Namely, the flame speed decreases to zero according to a linear law: c is proportional to $(\beta - \beta_e)$ for β close to β_e, in contrast to the quadratic dependence for $L_A < 1$. Similar results were obtained earlier in Gubernov et al. [2008a] analytically. Qualitatively the dependence of $c(\beta)$ is the same for the Lewis number for radicals varying over two orders of magnitude from 0.1 to 10.0 as shown in Gubernov et al. [2008b].

The dependence of the flame speed, c, on β becomes C-shaped for $L_A > 1$ i.e. $c(\beta)$ is a double-valued function. There are either two solutions travelling with different flame speed

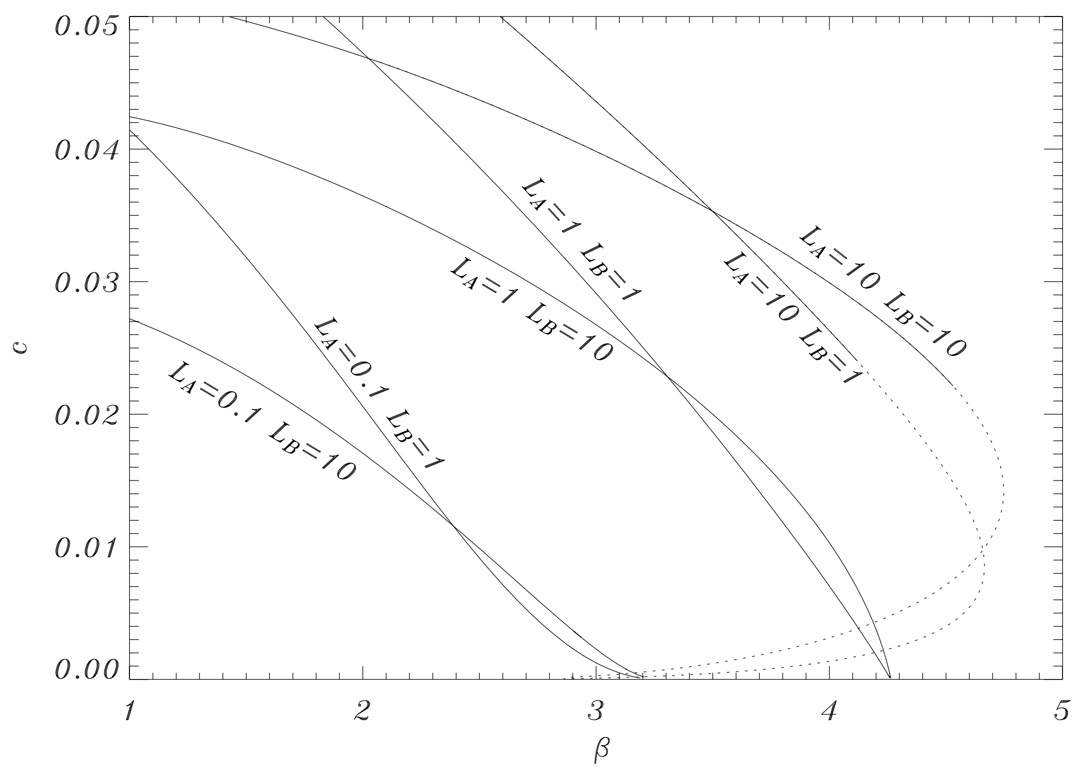

Figure 1: The dependence of the flame speed, c, on dimensionless activation energy, β, for $L_A = 0.1$, 1.0, 10.0, and two values of the Lewis number for radicals, $L_B = 1.0$ and $L_B = 10.0$. Solid lines represent the stable solutions, whereas unstable solutions are plotted with dotted lines.

or the solutions cease to exist due to the extinction when the fast solution branch meets the slow solution branch at the turning point of the $c(\beta)$ curve. We denote the coordinates of the turning point with subscript 'tp', i.e. β_{tp} and $c_{tp} = c(\beta_{tp})$. The dependence of the flame speed, c, on dimensionless activation energy, β, is shown in Fig. 1 for $L_A = 10$ and various values of L_B. In Fig. 1 the stable solution branches are plotted with the solid line and the dotted line represents the unstable branches (the stability analysis is described in detail in the next Sec.). For small values of β the fast solution branch is stable and is characterized by a negligibly small residual amount of fuel. As the activation energy is increased, the residual amount of fuel grows and it becomes significant as the turning point of $c(\beta)$ is approached. The slow solution branch is always unstable. It is characterized by a considerable fuel leakage. As we move along the slow solution branch by decreasing β from the turning point value, the flame speed decreases and at certain value β_e it becomes equal to zero. The behaviour of $c(\beta)$ follows a quadratic law i.e. $c \sim (\beta - \beta_e)^2$ as the dimensionless activation energy approaches the extinction value β_e. As shown in Gubernov et al. [2008b], variation of L_B over two orders of magnitude does not effect the qualitative behavior of the solution branches in the parameter space.

4 Stability of the Travelling Combustion Waves

In order to investigate the stability of combustion waves with respect to pulsating perturbations we linearize the governing Eqs. (1) near the travelling wave solution, that is we seek the solution of the form $u(\xi,t) = U(\xi) + \epsilon\phi(\xi)\exp(\lambda t)$, $v(\xi,t) = V(\xi) + \epsilon\psi(\xi)\exp(\lambda t)$, and $w(\xi,t) = W(\xi) + \epsilon\chi(\xi)\exp(\lambda t)$, where $[U(\xi),\ V(\xi),\ W(\xi)]$ represent the travelling combustion wave and terms proportional to the small parameter ϵ are the linear perturbation terms. Substituting this expansion into (1), leaving terms proportional to the first order of ϵ only, and introducing the vector function with components $\mathbf{v}(\xi) = [\phi,\ \psi,\ \chi,\ \phi_\xi,\ \psi_\xi,\ \chi_\xi]^T$ we obtain

$$\mathbf{v}_\xi = \hat{A}(\xi,\lambda)\mathbf{v}, \tag{6}$$

where

$$\hat{A} = \begin{bmatrix} 0 & \hat{I} \\ \hat{H}(\xi,\lambda) & \hat{C} \end{bmatrix}, \tag{7}$$

$$\hat{H} = \begin{bmatrix} \lambda & 0 & -r \\ \beta L_A \dfrac{VW}{U^2} e^{-1/U} & \beta L_A W e^{-1/U} + L_A\lambda & \beta L_A V e^{-1/U} \\ -\beta L_B \dfrac{VW}{U^2} e^{-1/U} & -\beta L_B W e^{-1/U} & -L_B(\beta V e^{-1/U} - \lambda - \beta r) \end{bmatrix}, \tag{8}$$

$$\hat{C} = \begin{bmatrix} -c & 0 & 0 \\ 0 & -cL_A & 0 \\ 0 & 0 & -cL_B \end{bmatrix}, \quad \hat{I} = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}. \tag{9}$$

We will call a set, Σ, of all λ values for which there exist a solution to (6) bounded for both $\xi \to \pm\infty$ a spectrum of linear perturbations. In the general case Σ is a set on a complex plane and it consists of the essential spectrum Σ_{ess} and the discrete spectrum Σ_{disc}. If there exists at least one $\lambda \in \Sigma$ such that $Re\lambda > 0$ then the travelling wave solution is linearly unstable, otherwise, if for all $\lambda \in \Sigma$ the real parts are not positive, then the travelling wave solution is linearly stable. Therefore in order to investigate the linear stability of the travelling wave solutions to (1), the spectrum Σ of the problem (6) has to be found. It can be shown (see Sandstede [2002] for details) that the essential spectrum is comprised

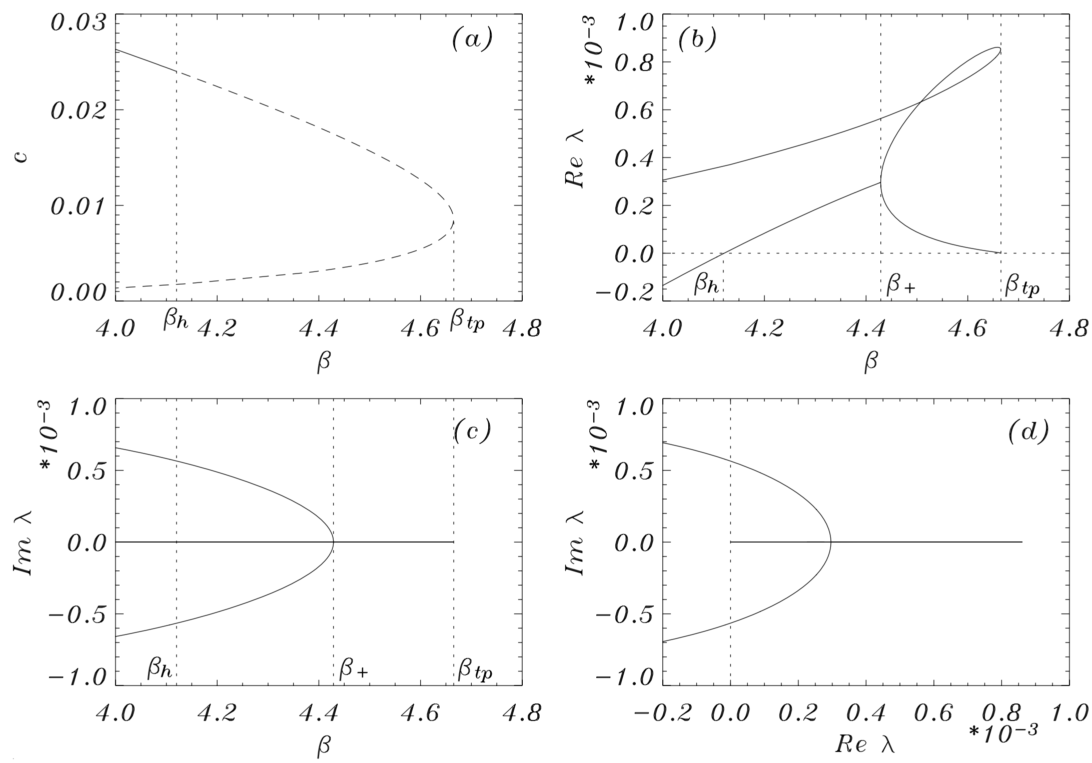

Figure 2: Bifurcation diagram for $L_A = 10.0$ and $L_B = 1.0$. The dependence of $c(\beta)$ is plotted in Fig. (a), where the solid line represents the stable and the dashed line unstable solutions. The real and imaginary parts of λ as functions of β are given in Figs. (b) and (c) respectively. In Fig. (d) the location of the points of the discrete spectrum as parameter β is varied in a complex plane is shown. In Figs. (a) - (c) β_h and β_{tp} represent the values of the parameter β when the Hopf bifurcation and fold bifurcation occur respectively and β_+ denotes the value of the parameter β when the points on the discrete spectra becomes real.

of parabolic curves on a complex plane with $Re\lambda \leq 0$. This implies that it is the discrete spectrum of the problem (6) that is responsible for the emergence of instabilities.

The linear stability problem is solved by finding the location of the discrete spectrum on a complex plane using the Evans function method [Evans, 1972] implemented with the use of a compound matrix approach [Gubernov et al., 2004]. However, in order to carry out such an investigation, the methods outlined in Gubernov et al. [2004] have to be generalized for a model with a two-step reaction mechanism. From the point of view of the compound matrix method the difference between the one-step model considered in Gubernov et al. [2004] and the current model with a two-step reaction mechanism is in the increased number of governing Eqs.. This substantially changes the topology of the linear stability problem in the compound matrix method formulation for the case of the two-step model and makes the methods described in Gubernov et al. [2004] not applicable to the case considered here. In order to overcome this difficulty we use an exterior algebra formulation of the Evans function method and generalize our approach to make it applicable to models with arbitrary number of governing Eqs. as described in Gubernov et al. [2006b].

The application of the Evans function method to the stability analysis allows one to obtain a location of the discrete spectrum on the complex plane and thus gives detailed information about pulsating instabilities emerging as a result of stability loss of the travelling combustion wave. Our analysis shows that for $L_A \leq 1$ and L_B varying over several orders of magnitude from 0.1 to 10.0 there are no points of the discrete spectrum located in the right half plane (i.e. $Re\lambda > 0$) and therefore the travelling wave solution is always stable with respect to pulsating instabilities. For the case $L_A > 1$ the travelling combustion wave is either stable or exhibits the onset of pulsations as a result of the Hopf bifurcation. This situation is illustrated in Fig. 2(a), where the dependence of the flame speed, c, on the dimensionless activation energy, β, is plotted for $L_A = 10.0$ and $L_B = 1.0$. The stable travelling wave solutions are represented by the solid line and the unstable solutions are plotted with the dashed line. The fast solution branch is found to be stable for small values of the activation energy. As β is increased towards the turning point a pair of the complex conjugate points of the discrete spectrum moves from the left ($Re\lambda < 0$) to the right ($Re\lambda > 0$) half of the complex plane crossing the imaginary axis with $Im\lambda \neq 0$ at a certain value $\beta = \beta_h$. This is illustrated with the dotted line marked as 'β_h' in Figs. 2 (b), (c), and (d), where the dependencies of $Re\lambda(\beta)$, $Im\lambda(\beta)$, and $Im\lambda(Re\lambda)$ are plotted respectively. The critical value $\beta_h \approx 4.120$ corresponds to the point of stability loss of the travelling wave solutions due to the emergence of the pulsating instabilities via the Hopf bifurcation. This is shown in Fig. 2 (b) as the graph $Re\lambda(\beta)$ crosses the line $Re\lambda = 0$ plotted with the horizontal dotted line. Further increase of β above the critical value β_h results in the increase of the real part of the pair of the points of the discrete spectrum as seen in Fig. 2 (b). At the same time the imaginary part of $\lambda \in \Sigma_{disc}$ decreases as shown in Fig. 2 (c) and at certain value of $\beta_+ \approx 4.429$ the points of the discrete spectrum merge together and become purely real as is clearly seen in Figs. 2 (c) and (d). This event marks the change of instability nature from pulsating to uniform. As β approaches the turning point $\beta_{tp} \approx 4.6648$ along the fast solution branch ($c > c_{tp} \approx 8.585 \cdot 10^{-3}$), one of the points of the discrete spectrum moves towards the origin and merges with it at $\beta = \beta_{tp}$ while the other one remains in the right half plane giving rise to uniform instability of the travelling combustion wave. The former point is shown in Fig. 2 (b) with the curve ending at the point ($Re\lambda = 0$, β_{tp}) and the latter corresponds to a loop in the dependence $Re\lambda(\beta)$ which is tangent to the vertical dotted line $\beta = \beta_{tp}$. As we pass the turning point of the $c(\beta)$ graph moving from the fast to the slow solution branch, one of the points of Σ_{disc} stays in the positive real axis, thus, the slow solution branch is unstable due to the uniform instability. The same scenario has been observed for L_B ranging from 0.1 to 10.0. It should be noted that similar scenario is reported in Gubernov et al. [2004] (see Fig. 7) and Gubernov & Kim [2006] for the case of one-step premixed and diffusion flames respectively.

Next step we investigate the stability of the travelling wave solutions with respect to pulsating instabilities for various values of the Lewis number for fuel, L_A, in order to obtain a detailed instability map and determine the character of bifurcations responsible for the onset of the pulsating instabilities. We fix the value of L_B to be equal to unity and vary L_A from 0.1 to 10.0. For each value of L_A the stability of the travelling combustion wave is investigated for a broad range of β values by finding the location of Σ_{disc} in the complex plane. The results of this study are summarized in Fig. 3 where the critical parameter values for the extinction and Hopf bifurcation are depicted with the solid and the dotted lines respectively on the L_A versus β plane for $L_B = 1$. The Lewis number for fuel is plotted in decimal logarithmic scale in order to map the range of its variation uniformly on the graph. The term extinction here needs further clarification. As the control parameters of the model (i.e. L_A, L_B, β) are varied, the travelling wave solution either ceases to exist as the flame speed decreases to zero for the case $L_A \leq 1$ or when the turning point of the $c(\beta)$ curve is reached for the case $L_A > 1$. Therefore, the extinction of the travelling wave solution occurs either as $c = 0$ for $L_A < 1$ or when $[dc/d\beta]^{-1} = 0$ for $L_A > 1$. In Fig. 3 the region below the solid line, marked as 'extinction' curve, corresponds to the parameter values where the travelling wave solutions do not exist. Therefore this region is marked with the 'no solutions' label. The parameter values above the extinction curve correspond to either the region where a single travelling wave exist $L_A \leq 1$ or to the region where two

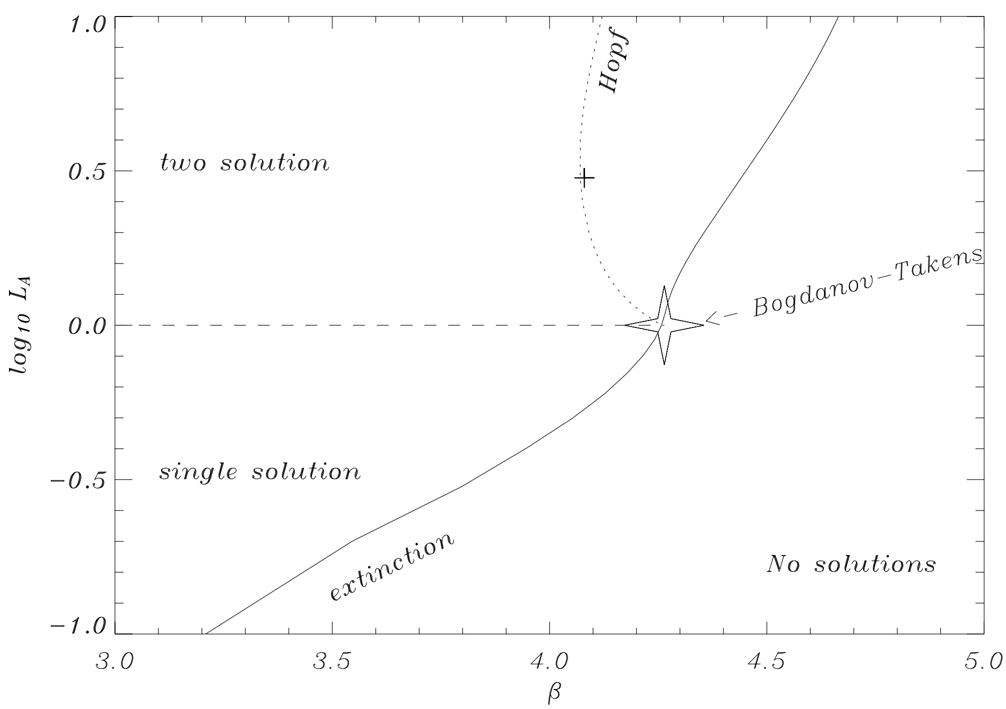

Figure 3: Stability diagram on the $(\beta, \log_{10} L_A)$, parameter plane for $L_B = 1$. The solid and dotted lines represent the loci of critical parameter values for extinction and Hopf bifurcation respectively. Sign '+' denotes the location of β and L_A chosen for the analysis presented in Fig. 5.

travelling wave solution branches may coexist $L_A > 1$. These two regions of parameters are marked with 'single solution' and 'two solution' labels and are separated by the dashed line $\log_{10} L_A = 0$ in Fig. 3. The stability analysis shows that for all parameters from the region below the $\log_{10} L_A = 0$ curve and above the extinction curve (single solution region in Fig. 3), the corresponding travelling wave solutions are stable (see Fig. 1). In contrast to this, in the parameter region where two solution branches may coexist the slow solution branch is always unstable and the fast solution branch is either stable or exhibits the onset of pulsating instabilities via the Hopf bifurcation. The dotted line corresponds to the Hopf bifurcation locus. Between the dotted (Hopf) and the solid line (extinction) lies a region where the fast travelling solution branch is unstable. Once the dotted line is crossed in the parameter space, for example, by increasing β for fixed L_A the fast branch of travelling wave solutions becomes unstable with respect to pulsating instabilities. Further increasing β and moving towards the extinction curve, the instability changes from pulsating to uniform as described above for the case $L_A = 10.0$.

Marked with a star on Fig. 3 is a point where the Hopf bifurcation curve meets the extinction curve. This point is a bifurcation of codimension two and is known as the Bogdanov-Takens bifurcation. It is a point from which the Hopf bifurcation locus in the parameter space originates and therefore this bifurcation is directly responsible for the onset of pulsations in the model. It is surprising that the Bogdanov-Takens bifurcation point is located on the line $L_A = 1$. Previously, we have shown the existence of this bifurcation for the case of both one-step premixed [Gubernov et al., 2004] and diffusion [Gubernov & Kim, 2006] flames. In these investigations the Bogdanov-Takens bifurcation is demonstrated to play an important role in the onset of pulsating instabilities. However, in the case of the one-step reaction the Bogdanov-Takens bifurcation point is found for Lewis numbers greater than one. This can have an important implication on the multidimensional stability which we will discuss later.

The stability diagram in the (β, L_A) parameter plane for various values of $L_B = 0.2$, 1.0, and 5.0 is plotted in Fig. 4. As is seen from Fig. 4 the variation of L_B does not effect the qualitative behaviour of the stability diagram, although the critical parameter values for the Hopf bifurcation substantially shifts towards the larger values of β with the increase in L_B. The extinction curve is only slightly influenced by variations in L_B, namely, the extinction curve rotates clockwise around the Bogdanov-Takens bifurcation point with the increase in L_B. It should be noted that the location of the Bogdanov-Takens bifurcation is not effected by variation in L_B .

5 Pulsating Solutions

We investigate the properties of pulsating combustion wave solutions emerging as a result of the Hopf bifurcation when the parameters reach critical values. The governing Eqs. (1) are solved in a sufficiently large coordinate domain with the boundary conditions (2) imposed at the edge points of the space grid. For our numerical algorithm we use the method of splitting with respect to physical processes. Initially we solve the set of ordinary differential Eqs. which describe the temperature and the species concentration variations due to the branching and recombination reactions by using the fourth order Runge-Kutta algorithm. As a next step, Eqs. of mass transfer for fuel and radicals are solved with the Crank-Nicholson method of the second order approximation in space and time. The initial conditions for the numerical scheme are taken in a form of the travelling wave solution (or autowave) of (3).

The results of our investigation are presented in Fig. 5, where the behaviour of pulsating combustion wave is illustrated for $L_A = 3.0$, $L_B = 1.0$, and $\beta = 4.08$. The value of β is taken above the critical value of dimensionless activation energy for the Hopf bifurcation, $\beta_h = 4.0703...$ This choice of parameters is shown in Fig. 3 with a thick cross located just

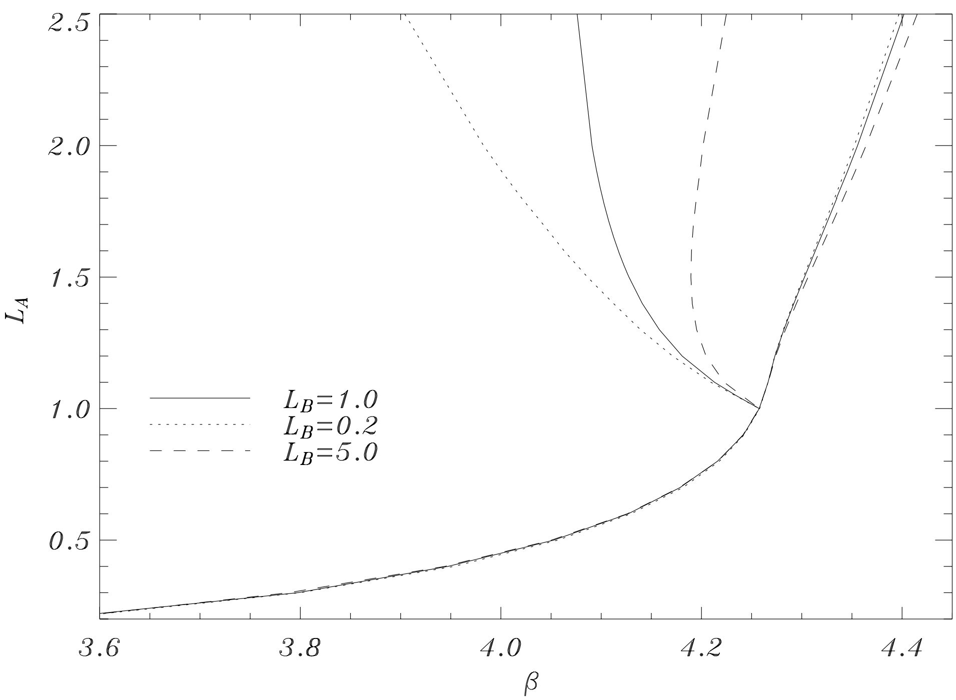

Figure 4: The Hopf bifurcation and extinction loci on the (β, L_A) parameter plane for $L_B = 1.0$, 0.2, and 5.0 with the regions defined as in Fig. 3.

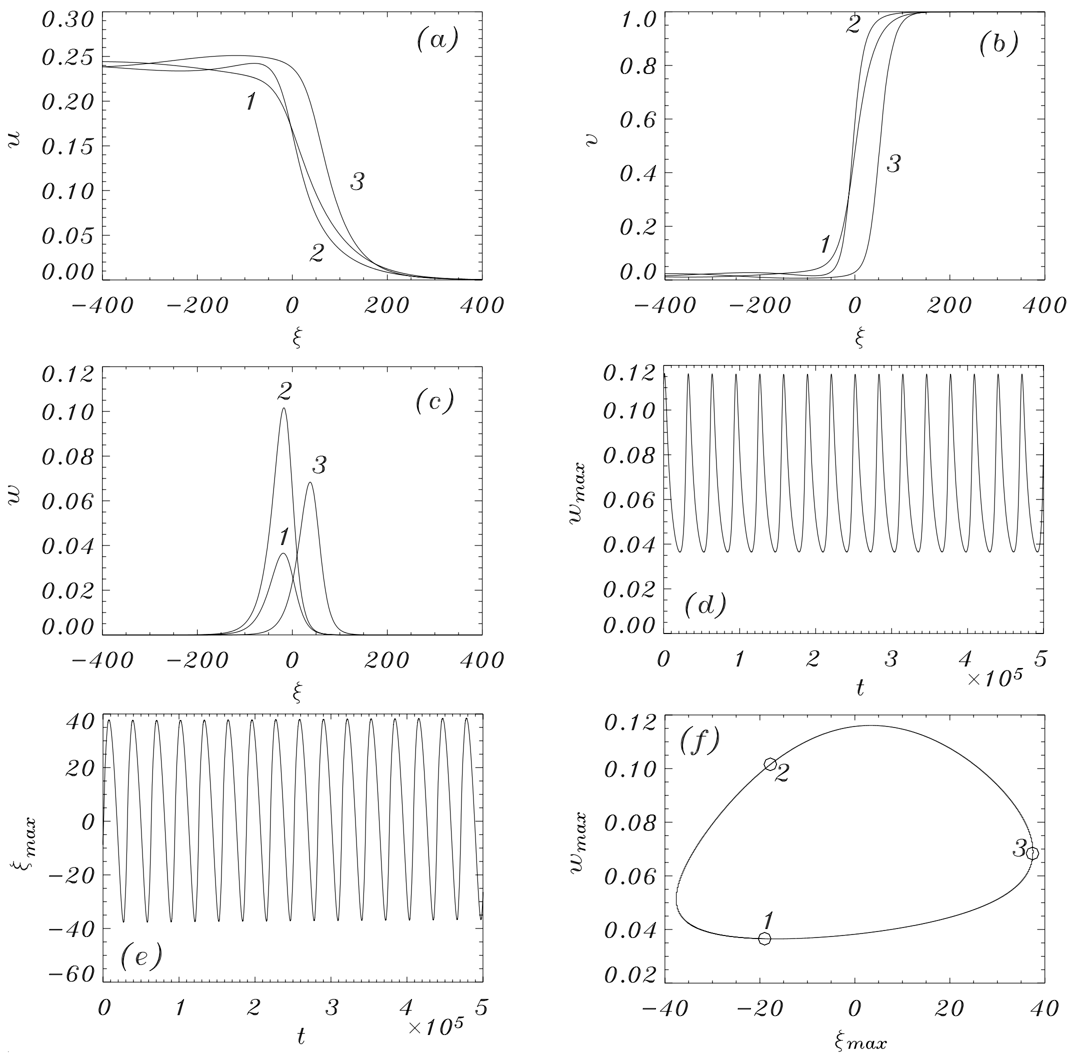

Figure 5: Pulsating combustion wave for $L_A = 3$, $L_B = 1$, and $\beta = 4.08$. Plotted in Figs. (a), (b), and (c) are temperature $u(\xi)$, concentration of fuel, $v(\xi)$, and concentration of radicals $w(\xi)$ profiles respectively. Solution profiles are sampled at $t_1 = 0$, $t_2 = 8750$, and $t_3 = 17500$ and are marked as 1, 2, and 3 respectively. The dependencies of instant values of w_{max} and ξ_{max} on time are presented in Figs. (d) and (e) respectively. In Fig. (f) the maximum value of the radicals concentration, w_{max}, is plotted againts the coordinate of the maximum, ξ_{max}.

beyond the Hopf curve (dotted line). The initial profile taken in the form of the travelling combustion wave is unstable and exhibits pulsating instabilities. These instabilities distort the solutions of (1) at the initial stages of the profile evolution in time. There are transient peaks in the temperature distribution in coordinate space and oscillations of the shape and maximum value of the radical concentration profile, w_{max}. The fuel concentration profile is mainly effected in the variation of the front curvature although some small oscillations of the fuel concentration are observed in the product region. The value of w_{max} and the location of the maximum of the radical concentration ξ_{max} are convenient parameters to describe the pulsating nature of the solution. Here $\xi = x - c_{drift}t$ is a coordinate in the frame travelling with the speed c_{drift} which is a mean value of the flame propagation $c_{drift} = \lim_{t\to\infty} x_{max}/t$, where x_{max} is a coordinate of the maximum of the radical concentration in the laboratory coordinate frame.

As the pulsating instabilities evolve, the value of w_{max} and ξ_{max} oscillate with an amplitude which grows exponentially with time. This type of behaviour is also reported in Gubernov et al. [2008b], where it is demonstrated that the frequency of these oscillations is given by the imaginary part $Im\lambda$ and the rate of exponential growth is determined by the real part, $Re\lambda$, of the pair of the points of the discrete spectrum, Σ_{disc}, responsible for the instability onset. However, at times of the order of $(Re\lambda)^{-1}$, the amplitudes of oscillations, $w_{max}(t)$ and $\xi_{max}(t)$, reach saturation and stabilize at certain values. The behaviour of $u(x,\ t)$, $v(x,\ t)$, and $w(x,\ t)$ profiles become periodic in time and the wave speed, defined as dx_{max}/dt, averaged over a period of pulsations is equal to c_{drift}, and so the pulsating combustion wave is formed.

In Fig. 5 (a), (b), and (c) the temperature, the concentration of fuel and the radicals profiles of the pulsating combustion wave are plotted respectively for three moments of time $t_1 = 0$, $t_2 = 8750$, and $t_3 = 17500$. Since the solution is periodic, time is measured from 0 to T, where $T \approx 30492.5$ is the period of oscillations. The coordinate $\xi = x - c_{drift}t$ is a coordinate in a travelling frame moving with speed c_{drfit}. In Fig. 5 (a) it is seen that the temperature profile, as a function of ξ, changes its behaviour over a period of time from monotonic for $t = t_1$ to a solution with a single local maximum as shown with curves 2 and 3. For $t = t_2$ the peak of temperature is relatively sharp, and for $t = t_3 > t_2$ it fades and the temperature profile gradually returns to a monotonic behaviour with respect to coordinate ξ. Also be noted that the instant coordinate of the maximum slope of $u(\xi)$ oscillates in time near $\xi = 0$. The fuel concentration profile $v(\xi,\ t)$ is plotted in Fig. 5 (b) for three successive moments of time. It is seen that the maximum slope of $v(\xi)$ for fixed t changes its value with time. Also the coordinate of the maximum slope exhibits oscillations over a period of time near the origin $\xi = 0$ in the travelling coordinate frame. It should be noted that since there is a residual amount of fuel left behind the combustion wave in the product region, some small oscillations of the fuel concentration leftovers are observed. The most remarkable soliton type dynamics is demonstrated by the radical concentration profile $w(\xi,\ t)$, which is depicted in Fig. 5 (c). As seen from this Fig. the radical concentration as a function of ξ remains a bell-shaped function of the soliton type for all moments of time. However, the maximum of $w(\xi)$ and its location are periodic functions of time. This is demonstrated in Figs. 5 (d) and (e), where the time dependencies of w_{max} and ξ_{max} are shown respectively. The distribution of radicals exhibits periodic oscillations near the average wave position $\xi = 0$. In Fig. 5 (f) $w_{max}(t)$ is plotted versus $\xi_{max}(t)$. It is seen that a limit cycle is formed in the $(\xi_{max},\ w_{max})$ plane.

To summarize, the pulsating combustion wave is travelling with an average speed, c_{drift}, however u, v, and w proflies exhibit periodic pulsations in time in such a way that the combustion wave accelerates and decelerates at certain moments of time and the flame propagates in a "stop-and-go" manner i.e. the wave accelerates, jumps to a new position, then decelerates and stops and so on. Similarly to that described above, the formation of pulsating waves after crossing the Hopf bifurcation curve in the parameter space has been

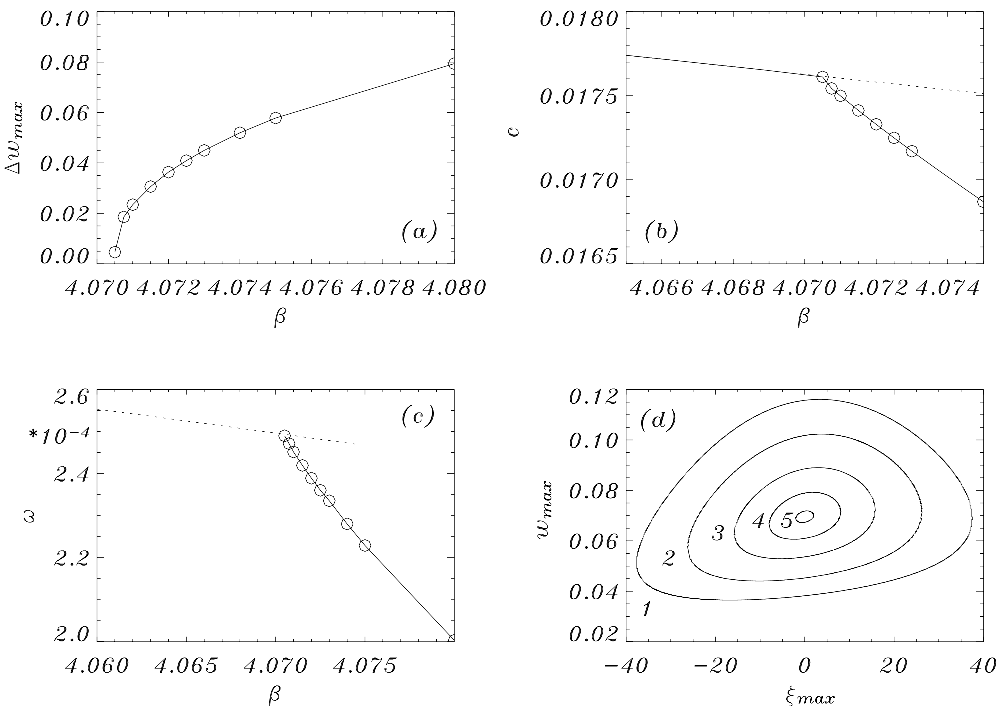

Figure 6: Hopf bifurcation and pulsating solution characteristics for $L_A = 3$ and $L_B = 1$. The dependence of the Δw_{max} amplitude of w_{max} oscillations on β is plotted in Fig. (a). In Fig. (b) the speed of the travelling combustion wave is plotted with the solid line for stable solution and with the dashed line for unstable solutions. The average drift speed of pulsating wave is plotted with the circles connected by the solid line. In Fig. (c) the frequency of oscillations, ω, is shown as a function of β with a solid line and the dashed line represents the imaginary part, $Im\lambda$, of the points of the discrete spectrum. In Fig. (d) $w_{max}(t)$ is plotted against $\xi_{max}(t)$ for various values of β (as described in the text).

observed for L_A ranging from 1 to 5. In Gubernov et al. [2008b] it is reported that no pulsating wave solutions are determined for $L_A = 10.0$. This implies that there could be a certain bifurcation for $5 < L_A < 10$ that changes the nature of the onset of pulsating instability. However, this issue has to be further clarified and lies beyond the scope of the current investigation.

In order to clarify the nature of the Hopf bifurcation, the properties of the pulsating solutions emerging from the travelling combustion wave have been investigated. The results of this studies are presented in Fig. 6, where all calculations have been undertaken for $L_A = 3.0$ and $L_B = 1.0$. Parameter β is varied from the values just above the critical value for the Hopf bifurcation $\beta_h = 4.0703...$ to $\beta = 4.08$ and the pulsating combustion wave is found by solving (1) for each β. In Figs. (a), (b), and (c) the circles correspond to numerical results, which are connected with the solid line. In Fig. 6 (a) the magnitude of oscillations of the maximum value of the radicals concentration, $\Delta w_{max} = \max\{w_{max}(t)\} - \min\{w_{max}(t)\}$ over one period $0 < t < T$, is plotted against β. The dependence of $\Delta w_{max}(\beta)$ shows root type behaviour typical for a periodic solution branch emanating from a Hopf bifurcation point. The Hopf bifurcation above is of supercritical type. In Fig. 6 (b) the speed of travelling combustion wave is plotted with the dashed line and the average drift speed is given by the circles connected with the solid line. As seen in Fig. 6 (b) the pulsating wave on average travels slower than the travelling wave and the difference $c - c_{drift}$ is growing almost linearly with increasing β. The frequency of oscillations of pulsating wave is presented in Fig. (c) with the circles connected with the solid line. It is also compared with the imaginary part of the pair of the points of the discrete spectrum responsible for the onset of instabilities for the travelling combustion wave which is plotted with the dashed line. It is seen that the frequency as well as flame speed is smaller for the pulsating than for the travelling wave. Finally, in Fig. 6 (d) the maximum value of the radicals concentration, $w_{max}(t)$, is plotted against $\xi_{max}(t)$ for several values of β: curve 1 corresponds to $\beta = 4.08$, curve 2 to $\beta = 4.075$, curve 3 to $\beta = 4.072$, curve 4 to $\beta = 4.07075$, and curve 5 to $\beta = 4.0705$. It is seen that on the $w_{max}(t)$ vs. $\xi_{max}(t)$ plane limit cycles are formed. The amplitude of oscillations grows as β increases. For $\beta = 4.0705$, just above the critical value for the Hopf bifurcation, the limit cycle has an almost elliptical shape and the $w_{max}(t)$ and $\xi_{max}(t)$ oscillations are close to harmonic. As β is increased the shape of the limit cycle deforms and becomes triangular for $\beta = 4.08$ indicating that $w_{max}(t)$ and $\xi_{max}(t)$ contain higher harmonics in a Fourier series expansion.

It should be noted that similar to the one-step models a period doubling bifurcation occurs with further increase of β. We have found both period two and period four solutions for the current model with chain branching reaction mechanism. These results are illustrated in Fig. 7, where $w_{max}(t)$ is given in (a, c) and $w_{max}(\xi_{max})$ is plotted in (b, d). the of period two solution is presented in Fig. 7 (a) and (b) for $\beta = 4.0823$. The limit cycle for $\beta = 4.08$ shown in Fig. 6 (d) consisted of a single closed curve, which splits into two loops for $\beta = 4.0823$ indicating the appearance of the period two solution. As β is further increased, a second period doubling bifurcation occurs and a solution of period four emerges. This period-four solution is illustrated in Figs. 7 (c, d) for $\beta = 4.0827$. The limit cycle for this case consists of four loops. We can expect that there exists a sequence of period doubling bifurcations leading chaotic solutions before extinction. A verification of this hypothesis is the subject future investigation.

6 Conclusions

We have investigated the properties and linear stability of the travelling premixed combustion waves and formation of the pulsating combustion waves in a model with two-step chain-branching reaction mechanism in the adiabatic limit. The model was introduced by Dold [2007]. The current paper naturally extends our previous analysis of the model in the equidiffusional approximation [Gubernov et al., 2006a; Gubernov et al., 2008a] and for the case of several fixed values of Lewis numbers for fuel and radicals [Gubernov et al., 2008b].

The investigation of the travelling combustion waves revealed that the Lewis number for fuel L_A has a substantial effect on the properties and the stability of the premixed flames in our model, whereas the variation of L_B effects only quantitatively the behaviour of the combustion waves. We have found that both the stability and the properties of the travelling combustion wave posses new and unique properties that have not been previously observed either for the one-step or for the two-step reaction models.

It was demonstrated that depending on the Lewis number for fuel the flame speed has either subcritical or supercritical behaviour as a function of the dimensionless activation energy. The transition from sub- to supercritical type of dependence occurs when $L_A = 1$. When the Lewis number for fuel is less than unity the flame speed is a single valued monotonically decreasing function of β. The flame extinction occurs as the speed of a combustion wave decays to zero in quadratic manner as a function of β when the critical values of the activation energy is approached i.e. $c \sim (\beta - \beta_e)^2$. For the Lewis number for fuel greater than unity the dependence of the flame speed on β is double-valued and C-shaped.

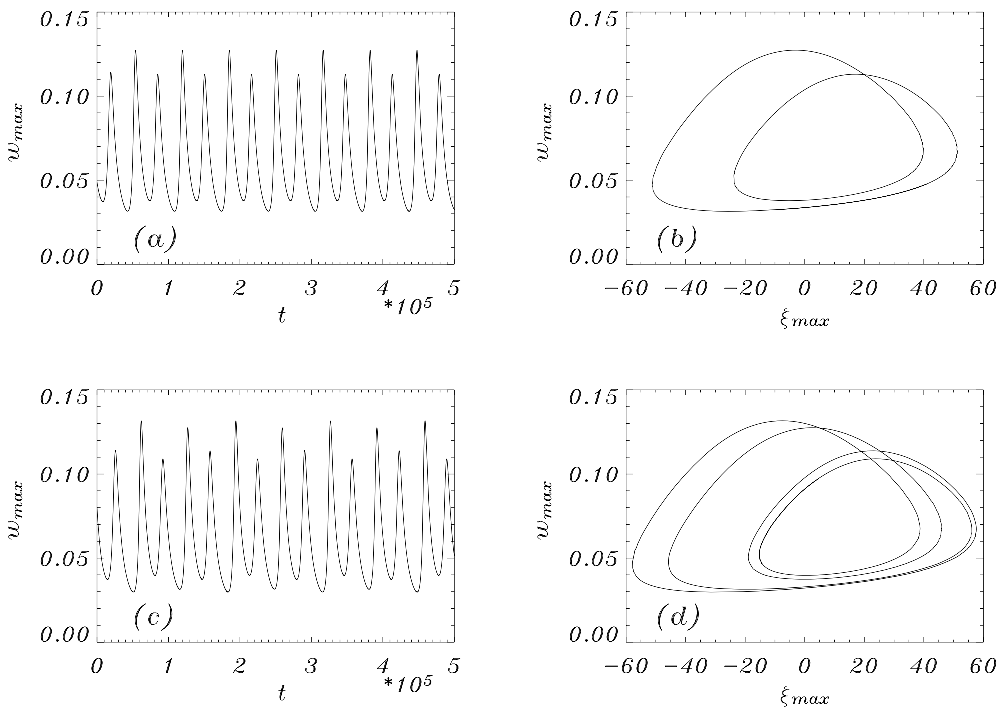

Figure 7: Period two and four pulsating solutions for $L_A = 3.0$, $L_B = 1.0$, $\beta = 4.0823$ and $\beta = 4.0827$ in Figs. (a-b) and (c-d) respectively. In Fig. (a) and (c) the dependence $w_{max}(t)$ is shown and in Fig. (b) and (d) the maximum value of the radicals concentration is plotted vs. ξ_{max}.

The travelling wave solutions either do not exist or there are two solutions travelling with different speed corresponding to fast and slow solution branches in the space of parameters. The flame extinction occurs when the slow and fast solution branches merge as a result of the turning point bifurcation. The slow solution branch is a monotonically increasing function of β. For larger values of β it ceases to exist at the turning point and for smaller β the flame speed decrease to zero as $c \sim (\beta - \beta_e)^2$ as the critical parameter value of the activation energy, β_e, is reached. For the fast solution branch $c(\beta)$ is a monotonically decreasing function. The transition from single to double valued character of the dependence of the flame speed on parameters occurs for the critical value of the Lewis number for fuel, $L_A = 1$. This case have been investigated earlier in Gubernov et al. [2006a] and Gubernov et al. [2008a] for $L_B = 1$ both numerically and analytically. It was shown that the speed of combustion wave is a unique monotonically decreasing function of β. The flame speed vanishes at the extinction point for finite value of activation energy, β_e, approaching zero according to linear law i.e. $c \sim (\beta - \beta_e)$.

The linear stability of the travelling combustion wave has been investigated in detail by using the Evans function method. The travelling combustion wave has been found to be stable with respect to pulsating instabilities for all parameter values for the case $L_A \leq 1$. For $L_A > 1$ the slow solution branch is always unstable and the fast solution branch is stable for small values of the dimensionless activation energy, β , and it loses stability via the Hopf bifurcation as β is increased towards the turning point. When the critical parameter values for the Hopf bifurcation are crossed in the space of parameters, a pair of complex conjugate points of the discrete spectrum moves from the left to the right half of the complex plane giving rise to the onset of pulsating instabilities. These instabilities can be characterized by a Hopf frequency which is determined by an imaginary part of the pair of unstable points of the discrete spectrum, Σ_{discr}, and also by a rate of growth which is determined by a real part of this pair of points. Both the critical parameter values for the Hopf bifurcation and the characteristics of emerging pulsating instabilities are determined for $1 < L_A < 10$ and various values of L_B ranging from 0.1 to 10. The Hopf bifurcation curve lies on the L_A vs β plane for $L_A > 1$ to the left from the extinction curve. As L_A is decreased from 10 to 1 both the distance between the Hopf bifurcation and the extinction locus as well as the Hopf frequency become smaller. At $L_A = 1$ the Hopf bifurcation and the extinction curve intersect, the Hopf frequency vanishes, and a bifurcation of codimension two takes place - a Bogdanov-Takens bifurcation. It is remarkable that the location of the Bogdanov-Takens bifurcation does not depend on the Lewis number for the radicals, L_B. This clearly shows that the case $L_A = 1$ is a distinguished limit, where the transition from one pattern in the flame dynamics to the other occurs. The Bogdanov-Takens bifurcation is responsible for both the change in the properties and stability of the travelling wave solution. At this point the extinction of the travelling wave solution is changing its behaviour from a subcritical to supercritical type. Furthermore since the Bogdanov-Takens bifurcation is a point in the parameter space where the Hopf bifurcation locus originates, it is responsible for the onset of pulsating instabilities. The results presented in this paper were calculated for $r = 0.001$. However, for r ranging over several orders of magnitude from 0.01 to 0.0001, the same qualitative behaviour of the properties and stability of the traveling combustion waves was found.

It is worthwhile noting that the cellular instabilities are known to originate from the extinction point at $L_A = 1$ for the case of one-step reaction models [Gubernov & Kim, 2006]. Therefore we can expect that for our current model the cellular instabilities may also appear from the Bogdanov-Takens bifurcation point and it is the bifurcation responsible for the onset of instabilities of all types. However, this issue requires a separate thorough investigation, which will be carried out in future work.

We have also investigated the properties of the Hopf bifurcation and the emerging pulsating combustion wave solutions for $1 < L_A < 5$. The direct integration of the governing partial differential Eqs. shows that the Hopf bifurcation is supercritical, the pulsations are excited in a so called soft regime and the magnitude of oscillations is growing continuously with the increase of $\beta - \beta_h$ in a root type manner. The properties of the pulsating waves have been investigated in detail. We have found that it is convenient to describe the dynamics of the pulsating waves by using w_{max} and ξ_{max} variables. On a plane of these variables each pulsating wave solution is represented by a limit cycle centered near the maximum value of radical concentration for the travelling combustion waves and $\xi_{max} = 0$. For small values of detuning of β from the critical value β_h, corresponding to the Hopf bifurcation, the limit cycle has an elliptical form indicating that the dynamics of pulsations is close to harmonic. As the detuning is increased the shape of the limit cycle deforms and becomes triangular. This implies that the higher order harmonics in the Fourier time series of dynamical variables become significant. It has been shown that on average the pulsating combustion wave propagates with a speed smaller than the speed of the travelling flame regime. The difference of the average drift speed of the pulsating wave and the corresponding speed of the travelling combustion wave decays almost linearly with increasing $\beta - \beta_h$. The frequency of pulsations also decays faster with increase of the dimensionless activation energy, than the Hopf frequency obtained from the linear stability analysis. We have considered so far moderate values of L_A only. In our previous study [Gubernov et al., 2008b] we were not able to find pulsating combustion wave solutions for $L_A = 10$. Our future investigation will clarify whether it is due to insufficient accuracy of the numerical calculations in Gubernov et al. [2008b] or there exists some other bifurcation responsible for the suppression of the pulsating solutions appearance for $L_A \sim 10$. From the analysis of the one-step reaction models it is expected that a period doubling bifurcation can emerge with further increase of detuning. We have observed this phenomenon for the current model with chain branching reaction mechanism. Pulsating solutions of period two and period four have been obtained. These solutions appear as a result of two sequential bifurcations of period doubling. It is not clear yet whether these bifurcations are part of the Feigenbaum period-doubling cascade leading to chaos. This issue needs clarification and will be investigated in the course of our ongoing work.

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