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**ON THE FAST-TIME CELLULAR INSTABILITIES  
OF LIÑÁN'S DIFFUSION-FLAME REGIME**

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## ON THE FAST-TIME CELLULAR INSTABILITIES OF LIÑÁN'S DIFFUSION-FLAME REGIME

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The fast-time instability of the Liñán's diffusion-flame regime is investigated asymptotically and numerically by employing the fast inner-zone time and length scales, as a model problem for the cellular instability in diffusion flames with Lewis numbers far from unity by an amount of order unity. The stability analysis revealed the full spectral nature, particularly near the saddle-node bifurcation condition corresponding to the minimum reduced Damköhler number  $\Delta$ . Contrary to the conventional belief, the minimum  $\Delta$  condition, commonly known as the Liñán's diffusion-flame extinction condition, is not necessarily an extinction condition for flames with Lewis numbers less than unity which can survive beyond the saddle-node bifurcation condition. The cellular instability could emerge upon passing the saddle-node bifurcation condition. The cellular instability is thus observable for near-extinction diffusion flames with Lewis numbers less than unity, as predicted by the previous experimental studies and the linear stability analysis employing the NEF limit. The stable parametric regions of small wave number and Lewis number just below unity were not predicted by the

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fast-time instability. But these parametric regions lie in the inner parametric layer of the distinguished limit employed in this analysis, so that the leading-order behavior is not contradictory with the previous experimental and analytical results. 30

*Keywords:* activation energy asymptotics, Liñán's diffusion-flame regime, fast-time instability

## INTRODUCTION

Because this volume of *Combustion Science and Technology* is a commemorative issue for Forman Williams's 70th birthday, this introduction is written from a perspective of the first author who is a former student of Forman Williams. I would like to begin this paper by mentioning how I stumbled into the problem of diffusion-flame instabilities. Of course, Forman Williams is a main player in this story. 35

It was the year 1993 when I was working for Forman Williams on the problem of acoustic instability in liquid propellant rocket engines. One day he handed me a paper on an experimental study on the diffusional-thermal instability in diffusion flames (Chen et al., 1992). The diffusional-thermal instability in premixed flames is well known to the combustion research community (Sivashinsky, 1977; Joulin and Clavin, 1979; Clavin, 1985), however that in diffusion flames had seldom been observed or only occasionally studied. Even though some previous publications on the subject (Garside and Jackson, 1953; Kirkby and Schmitz, 1966; Dongworth and Melvin, 1976; Ishizuka and Tsuji, 1981) exist, the findings in those papers are often fortuitous, so that the physical nature of the problem was hardly understood. It was only 1992 when Paul Ronney and his colleagues took a more systematic approach to experimentally investigate the diffusion-flame counterpart of the diffusional-thermal instability, and they found that periodic quenching marks could be formed in near-extinction diffusion flames with effective Lewis numbers much lower than unity. Their paper was presented in the 24th Symposium (International) on Combustion. Forman Williams of course did not forget to add a comment that it will require only a "simple" linear stability analysis to prove that the experimental findings are caused by the diffusional-thermal instability. The problem was handed down to me because during that time I was working on the acoustic response of diffusion flames (Kim and Williams, 1994), 40 45 50 55 60

that can be a subject as close to intrinsic flame instability as it can be. I started working on the problem since that moment, but it turned out to be that the stability analysis on diffusion-flame instability by the diffusional-thermal mechanism is anything but a “simple” linear stability analysis. Nevertheless, I am still very grateful that the problem was not simple. It still remains as one of the most important research topics of mine, and many problems that I am working on now have sprung from the problem. I could say that the problem of diffusional-thermal instability in diffusion flames is the backbone of my research career, and I must thank Forman Williams for giving me such an opportunity.

Before discussing the linear stability analysis, it is useful to point out that the activation energy asymptotics for diffusion flames has a crucial distinction from that of premixed flames. Because the leading-order solution to the diffusion-flame structure is the Burke-Schumann solution, effects of finite-rate chemical kinetics in diffusion flames first appear in the  $O(\beta^{-1})$  reactant leakage terms (where  $\beta$  denoting the Zel'dovich number), whereas those in premixed flames appear from the leading order. If the conventional near equidiffusional flame (NEF) limit (Buckmaster and Ludford, 1982), where deviation of the Lewis number from unity is only of  $O(\beta^{-1})$ , is adopted, finite-rate chemical-kinetic effects appear in  $O(\beta^{-2})$ . In order to avoid the situation carrying the analysis into that high order, Kim and Williams (1996) took a limit that the Lewis number is allowed to deviate from unity by an amount of order unity. However, the instability in that limit was so strong that the wavelength corresponding to the fastest growing mode was much shorter than the conventional convective-diffusive length scale, and the stability-analysis results obtained by using the length scales of the convective-diffusive layer and diffusive-reactive layer had to be composed to predict the properties of the cellular instability in diffusion flames. Eventually the linear stability analysis employing the NEF limit was carried out later (Kim, 1997) to obtain the instability results exhibiting its full nature in the convective-diffusive length scale only, even if the analysis was far more complex than its premixed flame counterpart.

For the distinguished limit of  $(L - 1) = O(1)$ , the unconventional analysis method employing the composite expansion led the results for the linear stability analysis to be somewhat awkward. However, the analysis still provides us with some valuable insights to the problem of diffusion-flame instability which might have been overlooked unless the problem is tackled in such an unorthodox manner. In particular,

the analysis revealed that the fast-time instability would be important for flames with Lewis number far from unity. It is the purpose of the present paper to shed more light on the problem of fast-time instability in diffusion flames because it is perhaps the most succinct form of the instability problem in diffusion flames and possesses all the necessary properties to explain the physical nature of the diffusional-thermal instability. 105

The fast-time instability was first considered by Peters (1978) to examine stability of the inner-zone structure of the Liñán's premixed flame regime. Then, the analysis was extended to diffusion flames by Q2 Buckmaster et al. (1982), who coined the terminology of the "fast-time instability" in the paper to emphasize the fact that the temporal coordinate is scaled by the fast inner-zone time scale. Later, those analyses were further extended to include the effects of three-dimensional reaction zone by Pereira and Vega (1990), of Lewis numbers greater than unity by Stewart and Buckmaster (1986), of Lewis numbers less than unity by Lozinski and Buckmaster (1995), and of the damping effect coming from the outer layer by Kim (1998). However, none of these papers are concerned with the full spectral characteristics of the fast-time instability. 110 115 120

This study presents the spectral characteristics of the fast-time instability for diffusion flames. Particular attention is focused on the cellular instability occurring with the fast-time scaling, so that the Lewis number will be restricted to be less than unity. Some of the spectral properties of the fast-time instability were examined when the cellular instability of diffusion flames was studied by Kim et al. (1996). However, the fast-time instability was not the main focus of the study, so that its complete nature was not properly emphasized and the asymptotic analysis was not in a better form. The present paper intends to fix the shortcomings in the previous results in order to provide a better overall picture of the fast-time instability for diffusion flames with Lewis numbers less than unity. The asymptotic analysis is further improved by introducing a more formal bifurcation technique, and its results are now better compared with the numerical solutions. In the present study, the fast-time instability is solved by the asymptotic method and numerical method. Near the bifurcation point, corresponding to the turning point of the Liñán's diffusion flame regime (Liñán, 1974), the asymptotic properties are derived and used as a guideline to the numerical analysis, employing the Evans function method (Sandstede, 2002). 125 130 135 140

In the following section, the governing equations for the mean and unsteady fields are introduced along with a brief description of the numerical method, and the mean field solution, which serves as the background solution to the instability problem, is presented in the third section. In the fourth section, the instability properties for planar disturbances are first examined by the asymptotic method to yield the condition of marginal stability and the growth behavior in the vicinity of the turning point. The asymptotic results are compared with the numerical results. The analysis is then extended to general wave numbers by using the numerical calculations. Based on the asymptotic results, obtained for the planar instability, an approximate dispersion relation and the corresponding non-linear evolution equation is derived. Finally, the concluding remarks and the future extensions to oscillatory instability and other activation energy asymptotic regimes are discussed in the fifth section.

## CONSERVATION EQUATIONS

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Since the conservation equations describing the inner structure of diffusion flames have been shown perhaps too many times elsewhere, the rather lengthy derivation, employing the activation energy asymptotics, is not presented here. The final equations for the mean field and unsteady field will be written here directly. The readers who wish to find the detailed derivation steps should refer to the previous paper by Kim et al. (1996).

### The Structure Equation for the Mean Field

The mean field inner structure is described by the famous Liñán's canonical equation for the diffusion flame regime:

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$$\frac{d^2\Theta}{d\xi^2} = \Delta(\Theta - \xi)(\Theta + \xi) \exp\{-(\Theta + \gamma\xi)\}$$

$$\Theta_\xi \rightarrow \pm 1 \quad \text{as} \quad \xi \rightarrow \pm\infty, \quad (1)$$

where  $\Theta$  is the inner variable for the temperature profile and  $\xi$  is the inner coordinate. The both variables stretched by the Zel'dovich number that is the primary expansion parameter for the activation energy asymptotics. The fuel and oxidizer concentrations in the inner layer are given by the coupling relationship as

$$\Theta_F = \Theta - \xi \quad \Theta_O = \Theta + \xi. \quad (2)$$

In the above mean field structure equation, the factor  $\gamma$  measures the degree of asymmetry in the thermal diffusion across the reaction zone. If  $\gamma = 0$ , the heat losses to both the fuel and oxidizer sides are equal. However, if  $\gamma$  is positive (negative), the heat loss to the oxidizer (fuel) side is greater. Therefore, the reaction in the oxidizer (fuel) side freezes faster and the fuel (oxidizer) leakage is expected to be greater than the oxidizer (fuel) leakage. For  $\gamma \rightarrow 1$  ( $\gamma \rightarrow -1$ ), the flame becomes nearly adiabatic to the oxidizer (fuel) side boundary, and the flame can sustain an extremely large fuel (oxidizer) leakage. Since the problem is symmetric to  $\gamma$ , we only need to solve for  $0 \leq \gamma < 1$ . In addition,  $\Delta$  is the reduced Damköhler number that is rescaled to be of order unity in the inner layer. For the notational brevity, the reduced Damköhler number  $\Delta$  will be just called Damköhler number unless the distinction from the unscaled Damöhler number is necessary. In the above equation, the mean-field problem is posed as that of finding the  $\Theta$  profile as a function of  $\Delta$ . It should be also kept in mind that the solution for  $\Theta$  is not necessarily a single valued function of  $\Delta$ .

### The Conservation Equations for the Linear Instability Analysis

In order to examine the stability of the inner flame structure, its time-dependent response on perturbations, imposed on the mean field solution, is considered. The differential equations describing the time-dependent behavior of an infinitesimally small normal-mode perturbation are written as

$$\begin{aligned} \frac{d^2\psi}{d\xi^2} - (S + K^2)\psi &= \frac{d^2\psi_F}{d\xi^2} - (LS + K^2)\psi_F = \frac{d^2\psi_O}{d\xi^2} - (LS + K^2)\psi_O \\ &= \Delta(\Theta - \xi)(\Theta + \xi)e^{-(\Theta+\gamma\xi)} \left[ \frac{\psi_F}{\Theta - \xi} + \frac{\psi_O}{\Theta + \xi} - \psi \right], \quad (3) \end{aligned}$$

where the Lewis number  $L$  is assumed to be identical for both the fuel and oxidizer. The growth rate  $S$  and the wave number  $K$  are scaled by the characteristic time and length of the inner diffusive-reactive layer, so that  $S$  and  $K$  are respectively two and one order of magnitude higher than their counterparts measured in the outer diffusive-convective layer.

Since the differential equations and the boundary conditions for  $\psi_F$  and  $\psi_O$  are identical as a consequence of the equal Lewis numbers of the two reactants, the fuel and oxidizer concentration perturbations have

a relationship  $\psi_F = \psi_O \equiv \chi(\xi)$ . Finally we have a simpler set of equations,

$$\frac{d^2\psi}{d\xi^2} - (S + K^2)\psi = \frac{d^2\chi}{d\xi^2} - (LS + K^2)\chi = \Delta e^{-(\Theta + \gamma\xi)} [2\Theta\chi - (\Theta^2 - \xi^2)\psi], \quad (4)$$

subject to  $\psi \rightarrow 0$  and  $\chi \rightarrow 0$  as  $\xi \rightarrow \pm\infty$ , from matching with the outer region where the perturbations are found to be vanishing at the leading order. This matching condition arises from the vanishing perturbations 210 outside of the inner layer because the perturbations are too fast and too short for them to survive in the outer layer. Because of this shorter time scale, instabilities found with this scaling often have been called fast-time instabilities (Buckmaster et al., 1983). The terms involving  $S$  represent the time-dependent effects in the reaction zone, and the 215 terms involving  $K^2$  account for the transverse diffusion in that zone, so that the reaction zone is neither planar nor quasi-steady any longer.

Even if the vanishing perturbation is the correct matching condition, it is often impractical to employ the strong boundary condition because it requires too big a calculation domain for the solution to converge to 220 the desired boundary condition. Therefore, we rather employ a weaker boundary condition given as

$$\frac{d\psi}{d\xi} \rightarrow \mp\sqrt{S + K^2}\psi, \quad \frac{d\chi}{d\xi} \rightarrow \mp\sqrt{LS + K^2}\chi, \quad \text{as } \xi \rightarrow \pm\infty, \quad (5)$$

which is obtained by expanding Eq. (4) for  $\xi \rightarrow \pm\infty$  and expresses the exponential decaying toward the boundary region. 225

In this stability problem, the Lewis number  $L$  and Damköhler number  $\Delta$  are the main control parameters. In this stability analysis, the problem is posed as that of finding the growth rate  $S$  as a function of the wave number  $K$  while the Lewis number  $L$  and the Damköhler number  $\Delta$  are specified. From these instability spectra, we seek to find the instability 230 characteristics of the diffusion-flame inner structure.

## Numerical Method

The key tool that is used for numerical investigation of stability of diffusion flames is the Evans function method (Sandstede, 2002). This method is relatively new for combustion science. Previously the Evans 235 function approach was employed to study the onset of pulsating instabilities in premixed flames with Lewis number  $L > 1$  (Gubernov et al.,



2003). In this paper, we extend the applicability of the method to investigate the instabilities of the different nature, namely, transversal or cellular instabilities in diffusion flames, which are dominant for the case of  $L < 1$ . 240

Prior to solving for the spectral problem, we solve the mean-field inner-zone problem numerically using shooting and relaxation methods. As a first step we solve Eq. (1) employing the fifth-order Runge-Kutta method in an interval  $\xi \in [-L_1, L_2]$ , where  $L_{1,2} > 0$  are taken to be sufficiently large. In our calculations  $L_{1,2}$  is chosen to keep the right-hand side of Eq. (1) to be less than  $10^{-20}$  for  $\xi \rightarrow -L_1$  or  $L_2$ . On the boundaries of the interval of integration, the reaction term (right-hand side) in Eq. (1) can be neglected and boundary conditions  $|\Theta_\xi| = 1$  can be used. Our numerical integrator allows us to estimate the local relative error, which has been set to be less than  $10^{-5}$  in our calculations. 245 250

As a second step the solution obtained with the shooting method is used as a guess solution for a more accurate method, namely relaxation. Here we would like to refer the reader to Gubernov et al. (2003) and references therein for the detailed description of the method. The stability analysis of the diffusion flames carried out in the following sections is based on the accuracy of our approximation of the solution to Eq. (1). The relaxation routine allows us to control the average local correction made on each iteration step. The solution is considered to be resolved if the correction is less than  $10^{-15}$ . 255 260

Once the mean-field solution is obtained, we solve for the linear stability problem numerically by using the Evans function method, which is described in detail in Gubernov et al. (2003). Employing the method described in the paper, the spectral problem in Eqs. (4) and (5) can be reduced to the search of zeroes of the Evans function  $D(S)$ , which has a very important property (Sandstede, 2002):  $D(S) = 0$  for some given value of  $S$  if and only if for this value of  $S$  Eq. (4) has at least one solution bounded for both  $\xi \rightarrow \pm\infty$  and satisfying the boundary conditions in Eq. (5). Consequently, we can look for zeroes of the Evans function, instead of solving linear stability problem in Eqs. (4) and (5) directly. In general both  $D$  and  $S$  are complex, however, in this paper we consider cellular instabilities only ( $L < 1$ ) and  $S$  can be taken to be a real number. Equation  $D(S) = 0$  is then solved numerically by using the Newton-Raphson method for fixed  $K$ . This enables us to determine the dispersion relation  $S(K^2)$  for any given parametric values of  $\gamma$ ,  $L$ , and  $\Delta$ . 265 270 275

## MEAN-FIELD SOLUTION

Even if the basic characteristics for the solution to Eq. (1) are already given by Liñán, the mean-field solution is presented here again because it serves as the base solution to the instability problem. In addition, its higher-order derivatives are essential to obtain the asymptotic properties of the spectrum to Eq. (4). 280

### General Characters of the Mean-Field Solutions

The overall characteristics of the mean-field solution could be better represented by the plot of the fuel leakage  $\alpha_F = \Theta_F(\xi \rightarrow \infty) \equiv (\Theta - \xi)(\xi \rightarrow \infty)$ . Here, only positive values of  $\gamma$  are considered because the problem is symmetric to the negative  $\gamma$  if  $\alpha_F$  is replaced by  $\alpha_O = \Theta_O(\xi \rightarrow -\infty) \equiv (\Theta + \xi)(\xi \rightarrow -\infty)$ . Therefore,  $\alpha_F$  will be simply denoted by  $\alpha$  for the notational brevity. The variation of  $\alpha$  with  $\Delta$  is shown in Figure 1, and it can be found that there exist minimum values of  $\Delta$ , below which solutions do not exist. The condition of minimum  $\Delta$  is 290

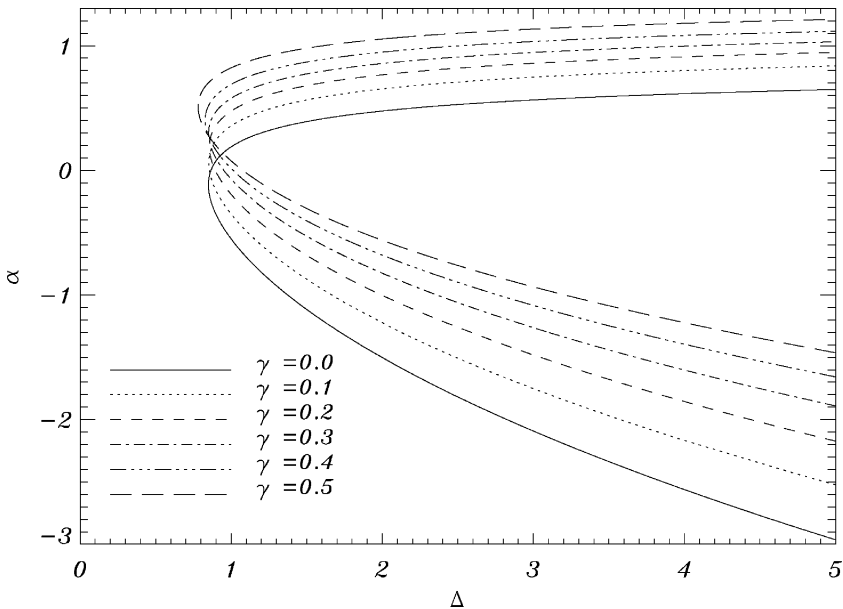


Figure 1. Dependence of the fuel leakage  $\alpha$  on the reduced Damköhler number  $\Delta$  for  $\gamma = 0.0, 0.1, 0.2, 0.3, 0.4$ , and  $0.5$ .

a saddle-node bifurcation point (or turning point), in the vicinity of which interesting dynamic behaviors are to be found, and our attention will be focused on analyzing the instabilities in this region. In the original paper by Liñán (1974), the minimum  $\Delta$  condition was identified as the first approximation to the extinction condition, and the condition is still widely used to identify the extinction conditions in diffusion flames. However, the condition of minimum  $\Delta$  does not necessarily correspond to the extinction condition if the Lewis number deviates from unity by an order of unity (Kim and Williams, 1997). For  $L < 1$ , the extinction condition is delayed beyond the saddle-node bifurcation point of Eq. (1) and the region between the saddle-node bifurcation point and the shifted extinction condition turns out to be the window of opportunity for the instabilities to arise.

The value of minimum  $\Delta$  as a function of  $\gamma$  is

$$\Delta_m = e\{(1 - |\gamma|) - (1 - |\gamma|)^2 + 0.26(1 - |\gamma|)^3 + 0.055(1 - |\gamma|)^4\}, \quad (6)$$

where the subscript  $m$  denotes the condition of minimum  $\Delta$  (Liñán, 1974). Contrary to the ordinary belief that the above equation is obtained by numerical fitting, it is an analytic correlation. The two-term expansion of  $\Delta_m$  can be obtained by the analytic procedure shown in the appendix (Clavin and Liñán, 1984). With an additional numerical result at  $\gamma = 0$ , the above four-term correlation is obtained, which is found to be in excellent agreement with the numerical data throughout the entire range of  $\gamma$  as shown in Figure 2.

As  $\gamma$  approaches unity, the fuel supply side becomes adiabatic, so that the flame tends to stay alive until the fuel leakage becomes extremely large. Under this circumstance, the inner structure resembles that of premixed flame and the so-called premixed-flame regime arises. Since this inner structure falls into a different distinguished limit, our attention will be rather restricted to the case where  $\gamma$  is close to zero. On the other hand, the instability properties near  $\gamma = 1$  will be left as a future work.

### Higher-Order Derivatives of the Inner Solutions

In order to obtain the asymptotic properties of the instability spectrum  $S(K^2)$ , it is essential to find the higher-order properties of Eq. (1). The method to obtain such properties, including the first- and second-order derivatives of the inner structure with respect to the fuel leakage, is presented here. In principle, the higher-order derivatives may be

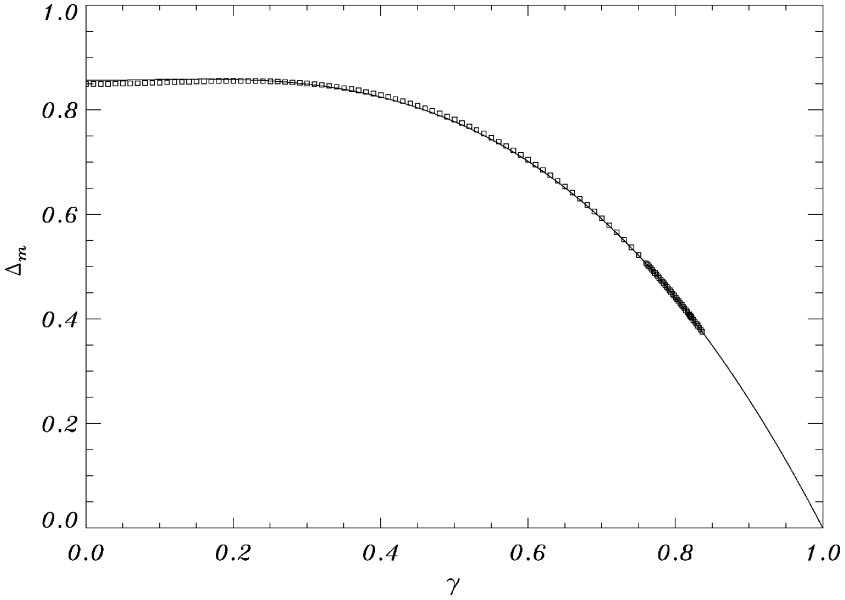


Figure 2. Dependence of  $\Delta_m$  on  $\gamma$ . The solid line is plotted according to Eq. (6), whereas the squares represent the results obtained numerically.

obtained by solving the problem defined in Eq. (1) for several values of  $\alpha$  and then differentiating numerically. However, this procedure is quite time-consuming but not sufficiently accurate. An alternative procedure 330 is thus employed here.

The alternative procedure to calculate the derivatives with respect to the fuel leakage involves expansion of Eq. (1) arising from small increment of the fuel leakage, denoted by  $\alpha_1$ , about a given value of the fuel leakage  $\alpha_0$ . Since  $\Theta$  and  $\Delta$  are parametrically dependent on  $\alpha$ , we 335 may have the expansions in the form

$$\left. \begin{aligned} \alpha &= \alpha_0 + \alpha_1, \\ \Theta(\xi; \alpha) &= \Theta(\xi; \alpha_0) + \frac{\partial \Theta}{\partial \alpha}(\xi; \alpha_0) \alpha_1 + \dots = \Theta(\xi; \alpha_0) + \vartheta \alpha_1 + \dots, \\ \Delta(\alpha) &= \Delta(\alpha_0) + \frac{d\Delta}{d\alpha} \Big|_{\alpha=\alpha_0} \alpha_1 + \dots = \Delta_0(1 + \Delta' \alpha_1 + \dots), \end{aligned} \right\} \quad (7)$$

where

$$\vartheta \equiv \frac{\partial \Theta}{\partial \alpha} \Big|_{\alpha=\alpha_0}, \quad \Delta' \equiv \frac{d \ln \Delta}{d \alpha} \Big|_{\alpha=\alpha_0}, \quad \Delta_0 \equiv \Delta(\alpha_0) \quad (8)$$

Substituting the above expansions into Eq. (1) and collecting the terms at order  $\alpha_1$  alone, we find the problem for determining  $\vartheta$  to be

$$\left. \begin{aligned} \frac{d^2 \vartheta}{d\xi^2} &= \Delta_0 e^{-(\Theta + \gamma \xi)} [(2\Theta - \Theta^2 + \xi^2)\vartheta + \Delta'(\Theta^2 - \xi^2)], \\ \frac{d\vartheta}{d\xi} &\rightarrow 0, \quad \text{as } \xi \rightarrow \pm\infty. \end{aligned} \right\} \quad (9)$$

To assure that the matching condition for the fuel leakage,  $\alpha = (\Theta - \xi)|_{\xi \rightarrow \infty}$ , is satisfied, a supplementary condition to Eq. (9) must be appended

$$\alpha_0 + \alpha_1 = \Theta(\xi; \alpha_0) + \vartheta \alpha_1 - \xi, \quad \text{as } \xi \rightarrow \infty. \quad (10)$$

Since  $\alpha_0 = (\Theta - \xi)|_{\xi \rightarrow \infty}$ , the applicable boundary condition for Eq. (9) is then found to be

$$\vartheta \rightarrow 1 \quad \text{as } \xi \rightarrow \infty. \quad (11)$$

Solution to Eq. (9) with the supplementary condition in Eq. (11) yields a unique function  $\vartheta$ , eigenvalue  $\Delta'$  and a constant value for  $\vartheta(-\infty)$  corresponding to  $r = d\alpha_O/d\alpha_F$ , required in the dispersion relation. For  $\gamma = 0$ ,  $r = 1$  for all values of  $\alpha$  thanks to the symmetry, and the corresponding numerical results for  $\Delta'$  are shown in Figure 3. In the figure, one should note that  $\Delta'$  (or  $\alpha$ ) is a single value function while moving along the solution branch shown in Figure 1. Therefore, it is desirable to express the mean field and spectrum as a function of  $\Delta'$  (or  $\alpha$ ) instead of  $\Delta$ .

Since  $\Delta'$  is zero at the saddle-node bifurcation point, the second-order derivative is needed in order to express  $\Delta'$  in the region while carrying out the bifurcation analysis. Here, we seek the solution in the form,

$$\left. \begin{aligned} \alpha &= \alpha_m + \alpha_1, \\ \Theta(\xi; \alpha) &= \Theta(\xi; \alpha_m) + \frac{\partial \theta}{\partial \alpha}(\xi; \alpha_m) \alpha_1 + \frac{\partial^2 \theta}{\partial \alpha^2}(\xi; \alpha_m) \frac{\alpha_1^2}{2} + \dots \\ &= \Theta(\xi; \alpha_m) + \vartheta_0 \alpha_1 + \vartheta_1 \frac{\alpha_1^2}{2} + \dots, \\ \Delta(\alpha) &= \Delta(\alpha_m) + \frac{d\Delta}{d\alpha} \Big|_{\alpha=\alpha_m} \alpha_1 + \frac{d^2 \Delta}{d\alpha^2} \Big|_{\alpha=\alpha_m} \frac{\alpha_1^2}{2} + \dots \\ &= \Delta_m (1 + \Delta'_m \alpha_1 + \Delta''_m \frac{\alpha_1^2}{2} + \dots), \end{aligned} \right\} \quad (12)$$

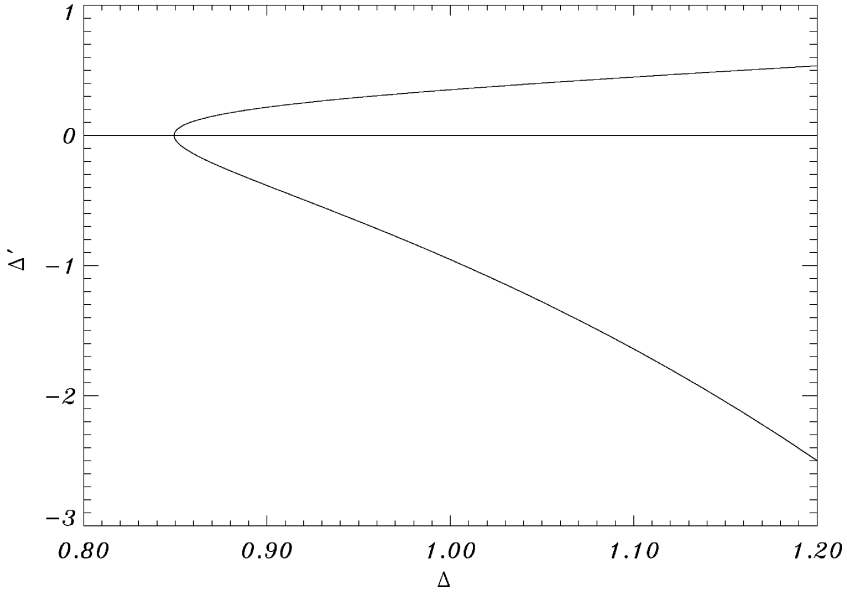


Figure 3. Dependence of  $\Delta'$  on  $\Delta$  for  $\gamma = 0$ .

where  $\Delta'_m = 0$  and

$$\Delta''_m \equiv \frac{1}{\Delta_m} \left. \frac{d^2 \Delta}{d\alpha^2} \right|_{\alpha=\alpha_m}$$

Substituting the above expansion into Eq. (1), the conservation equations for the first two orders of expansion are found to be

$$\mathcal{L}\vartheta_0 = 0 \tag{13}$$

$$\mathcal{L}\vartheta_1 = V_2\vartheta_0^2 + V_0\Delta''_m \tag{14}$$

where the linear differential operator  $\mathcal{L}$  and the potential functions  $V_i(\xi)$  are given as

$$\begin{aligned} \mathcal{L} &= d_{\xi\xi}^2 - V_1(\xi) \\ V_0(\xi) &= \Delta_m e^{-(\Theta+\gamma\xi)} (\Theta^2 - \xi^2) \\ V_1(\xi) &= \Delta_m e^{-(\Theta+\gamma\xi)} (2\Theta - \Theta^2 + \xi^2) \\ V_2(\xi) &= \Delta_m e^{-(\Theta+\gamma\xi)} (2 - 4\Theta + \Theta^2 - \xi^2) \end{aligned} \tag{15}$$

Obviously,  $\vartheta_0$  turns out to be the derivative of  $\Theta$  with respect to  $\alpha$  at the turning point, and its solution can be obtained by integrating Eq. (9) with the constraint  $\Delta' = 0$ . At order of  $\alpha_1^2$ , we have the inhomogeneous term  $V_2\vartheta_0^2 + V_0\Delta_m''$  and solution to Eq. (14) exists only if the projection of the inhomogeneous term to the homogeneous eigenfunction  $\vartheta_0$  vanishes. Consequently, we have

$$\langle V_2, \vartheta_0^3 \rangle + \langle V_0, \vartheta_0 \rangle \Delta_m'' = 0 \quad (16)$$

Therefore, the curvature of  $\Delta - \alpha_F$  curve becomes 380

$$\Delta_m'' = -\frac{\langle V_2, \vartheta_0^3 \rangle}{\langle V_0, \vartheta_0 \rangle} \quad (17)$$

The value of  $\Delta_m''$  at  $\gamma = 0$  is found to be 0.8220. In the limit of  $\gamma \rightarrow \pm 1$ , the asymptotic expression for  $\Delta_m''$  is also given as (Kim, 1997)

$$\Delta_m'' = \frac{(1 - |\gamma|)^2}{2} \quad (18)$$

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## SPECTRAL SOLUTION TO THE FAST-TIME INSTABILITY

Once the mean-field solution is obtained, the linear stability of external disturbances can be examined. In principle, the spectral problem in Eq. (4) with the boundary condition in Eq. (5) could be solved numerically. However, we may find some local asymptotic properties of the spectrum particularly in the neighborhood of the saddle-node bifurcation point and the asymptotic solutions then serve as the guideline to the numerical simulations. Comparison of the asymptotic solutions with the numerical solutions will certainly provide a better understanding of the spectral problem. In this paper, we will focus our attention first to the instabilities for planar disturbances and then move to the corresponding behaviors for disturbances with finite wave numbers. 390  
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### Instabilities for Planar Disturbances

*Onset Condition of the Fast-Time Instability for Planar Disturbances.* The eigenvalue problem describing the fast-time instability for 400

$K = 0$  with general Lewis number can be written from Eq. (4) as

$$\left. \begin{aligned} \frac{d^2\psi}{d\xi^2} - S\psi &= \frac{d^2\chi}{d\xi^2} - LS\chi = \Delta e^{-(\Theta+\gamma\xi)} [2\Theta\chi - (\Theta^2 - \xi^2)\psi], \\ \frac{d\psi}{d\xi} &\rightarrow \mp\sqrt{S}\psi, \quad \frac{d\chi}{d\xi} \rightarrow \mp\sqrt{LS}\chi \quad \text{as } \xi \rightarrow \pm\infty. \end{aligned} \right\} \quad (19)$$

Here we are seeking the condition at which the largest eigenvalue  $S$  begins to be positive. 405

Considering a situation immediately after the onset of planar instability,  $S$  is an infinitesimally small positive number. If  $\sqrt{S}$  is employed as a small expansion parameter, the eigenvalue problem becomes, at the leading order,

$$\frac{d^2\psi}{d\xi^2} = \frac{d^2\chi}{d\xi^2} = \Delta e^{-(\Theta+\gamma\xi)} [2\Theta\chi - (\Theta^2 - \xi^2)\psi], \quad (20)$$

which now has a coupling function for  $\psi - \chi$  in a linear functional form. The slope and integration constant for the coupling function is obtained by imposing the matching condition. In order to achieve matching, it must be noted that decay of the solution to zero takes place in an outer region with thickness of order  $1/\sqrt{S}$  in the  $\xi$  coordinate. In terms 415 of the coordinate  $\zeta = \sqrt{S}\xi$ , the differential equations in the outer layer become

$$\frac{d^2\psi}{d\zeta^2} - \psi = 0, \quad \frac{d^2\chi}{d\zeta^2} - L\chi = 0. \quad (21)$$

The exponentially decaying outer solutions are then found to be

$$\psi = \begin{cases} \psi^+ e^{-\zeta} \\ \psi^- e^{\zeta}, \end{cases} \quad \chi = \begin{cases} \chi^+ e^{-\sqrt{L}\zeta} \\ \chi^- e^{\sqrt{L}\zeta}, \end{cases} \quad \text{for } \begin{cases} \zeta > 0 \\ \zeta < 0, \end{cases} \quad (22)$$

where the integral constants  $\psi^\pm$  and  $\chi^\pm$  are yet to be determined. In the double limit of  $\zeta \rightarrow \pm 0$  and  $\xi \rightarrow \pm\infty$ , matching is achieved to yield  $\psi^+ - \psi^- = \chi^+ - \chi^-$  and  $\psi^+ + \psi^- = \sqrt{L}(\chi^+ + \chi^-)$ , and a unique coupling function at leading order is found to be

$$\psi - \chi = (\sqrt{L} - 1) \frac{\chi^+ + \chi^-}{2}. \quad (23)$$

The value of  $\chi^+$  can be chosen arbitrarily because the problem is linear and homogeneous. With the choice of  $\chi^+ = 1$ , the resulting eigenvalue



problem, written in terms of  $\chi$ , becomes

$$\left. \begin{aligned} \frac{d^2\chi}{d\xi^2} &= \Delta e^{-(\Theta+\gamma\xi)} [(2\Theta - \Theta^2 + \xi^2)\chi + \frac{1+\chi^-}{2}(1-\sqrt{L})(\Theta^2 - \xi^2)], \\ \chi &\rightarrow 1 \quad \text{as } \xi \rightarrow \infty, \quad \chi \rightarrow \chi^- \quad \text{as } \xi \rightarrow -\infty. \end{aligned} \right\} \quad (24)$$

Comparison of this equation with Eqs. (9) and (11) shows that  $\chi^- = r$  430 and that an eigensolution exists if  $\Delta' = (1 - \sqrt{L})(1 + r)/2$ . Although the eigenfunction  $\chi$  must approach zero as  $\xi \rightarrow \pm\infty$ , that of Eq. (24) approaches nonzero constant values,  $\chi|_{\xi \rightarrow \infty} \rightarrow 1$  and  $\chi|_{\xi \rightarrow -\infty} \rightarrow r$ . The composite expansion of the inner and exponentially decaying outer solutions, however, provides an eigenfunction that is uniformly valid 435 throughout the entire range of  $\xi$  and that exhibits the correct exponential decay at infinity. Since this eigensolution exists only near the onset of instability, the onset condition of fast-time instability for the planar wave is

$$\Delta' = (1 - \sqrt{L}) \frac{1+r}{2}. \quad (25)$$

If the Lewis number is close to unity, the above onset criterion can be derived by a formal bifurcation analysis. Since the onset of instability occurs in the vicinity of the saddle-node bifurcation point, the following expansions are introduced:

$$\left. \begin{aligned} L &= 1 + \varepsilon, \\ \alpha &= \alpha_m + \varepsilon\alpha_1, \\ \Delta &= \Delta_m(1 + \varepsilon^2 \Delta_m'' \frac{\alpha_1^2}{2} + \dots), \\ \Theta &= \Theta_m + \varepsilon\alpha_1 \Theta_1 + \dots \\ \chi &= \chi_m + \varepsilon\alpha_1 \chi_1 + \dots \end{aligned} \right\} \quad (26)$$

Substituting the above expansions into Eq. (24), we find at the leading order

$$\left. \begin{aligned} \frac{d^2\chi_m}{d\xi^2} &= \Delta_m e^{-(\Theta_m+\gamma\xi)} [(2\Theta_m - \Theta_m^2 + \xi^2)\chi_m], \\ \chi_m &\rightarrow 1 \quad \text{as } \xi \rightarrow \infty, \quad \chi_m \rightarrow \chi^- \quad \text{as } \xi \rightarrow -\infty. \end{aligned} \right\} \quad (27)$$

Then the leading order solution to  $\chi_m$  is simply found to be

$$\chi_m = \left. \frac{d\Theta}{d\alpha} \right|_{\alpha_m} = \vartheta_m \tag{28}$$

and  $\chi^- = r_m$  where  $r_m$  is the value of  $r$  at the turning point.

Collecting the terms of  $O(\varepsilon)$ ,

$$\mathcal{L}\chi_1 = V_2\Theta_1^2 - V_0 \frac{1+r_m}{4\alpha_1} \tag{29}$$

where the linear differential operator  $\mathcal{L}$  is identical to that in Eq. (15). Again applying the solvability condition to the above equation, we find, 455

$$\langle V_2, \vartheta_0^3 \rangle - \langle V_0, \vartheta_0 \rangle \frac{1+r_m}{4\alpha_1} = 0 \tag{30}$$

where uses have been made of the identities  $\Theta_1 = \chi_m = \vartheta_0$  and  $\langle V_2, \vartheta_0^3 \rangle = -\langle V_0, \vartheta_0 \rangle \Delta_m''$ . Then we have the onset condition of

$$-\frac{1+r_m}{4\alpha_1} = \Delta_m'' \tag{31}$$

Since  $\Delta' = \Delta_m'' \varepsilon \alpha_1$ , the onset condition can be written in terms of  $\Delta'$  as 460

$$\Delta' = -\frac{1+r_m}{4} \varepsilon \tag{32}$$

that corresponds to the linear expansion of Eq. (25) for the limit of  $L \rightarrow 1$ .

*Growth Rate S with  $L = 1$  Near the Saddle-Node Bifurcation Point.* For  $L = 1$ , planar disturbances become unstable as soon as the 465 turning point is passed. Here we try to obtain the growth rate for planar disturbances just past the turning point. The corresponding differential equation is

$$\frac{d^2\psi}{d\xi^2} - \Delta e^{-(\Theta+\gamma\xi)} [(2\Theta - \Theta^2 + \xi^2)]\psi - S\psi = \mathcal{L}\psi - S\psi = 0 \tag{33}$$

where the linear differential operator is again given as  $\mathcal{L} \equiv d_{\xi\xi}^2 - V_1(\xi)$ . 470

In the region close to the saddle-node bifurcation point, we employ  $\delta = \alpha - \alpha_m$  as a small expansion parameter. Then, the variables are

expanded as

$$\left. \begin{aligned} \Delta &= \Delta_m(1 + \delta^2 \frac{\Delta_m''}{2} + \dots), \\ S &= \delta^2 \Sigma + \dots, \\ \psi &= \psi_m + \delta \psi_1 + \dots, \\ \Theta &= \Theta_m + \delta \vartheta_m + \dots, \end{aligned} \right\} \quad (34)$$

where  $\vartheta_m = (\partial\Theta/\partial\alpha)|_{\Delta'=0}$  and  $\Delta_m'' = (\Delta_m^{-1} d^2\Delta/d\alpha^2)|_{\Delta'=0}$  475

When these expansions are substituted into Eq. (33), the problem at order unity becomes

$$\frac{d^2\psi_m}{d\xi^2} - V_1(\xi)\psi_m = \mathcal{L}\psi_m = 0, \quad \psi_{m,\xi} \rightarrow 0 \quad \text{as } \xi \rightarrow \pm\infty, \quad (35)$$

Then, the solution to  $\psi_m$  is found to be

$$\psi_m = \vartheta_m \quad (36)$$

with  $\psi_{m,\infty} = 1$  and  $\psi_{m,-\infty} = r_m$ .

Collecting the terms of  $O(\delta)$ , the differential equation for  $\psi_1$  becomes

$$\mathcal{L}\psi_1 = V_2\vartheta_m^2 \quad (37)$$

where use has been made of the relationship  $\psi_m = \vartheta_m$ . However, care 485  
must be taken in determining the boundary condition to the above equation because there is a slowly decaying outer-layer solution. For the stretched coordinate

$$\zeta \equiv \xi/\delta$$

the slowly decaying problem in the outer layer, corresponding to Eq. (33), 490  
becomes

$$\frac{d^2\psi_{out}}{d\zeta^2} - \Sigma\psi_{out} = 0 \quad (38)$$

where the subscript *out* denotes the outer layer. The boundary conditions to Eq. (38) are found from the matching condition with the leading-order inner solution as 495

$$\psi_{out}(\infty) = 0 \quad \psi_{out}(0^+) = 1 \quad \psi_{out}(0^-) = r_m \quad \psi_{out}(-\infty) = 0 \quad (39)$$

Therefore, the solution for  $\psi_{out}$  becomes

$$\psi_{out} = \begin{cases} \exp(-\Sigma^{1/2}\zeta) & \text{for } \zeta > 0 \\ r_m \exp(\Sigma^{1/2}\zeta), & \zeta < 0, \end{cases} \quad (40)$$

Matching with this outer decaying solution provides the boundary condition to Eq. (37) as

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$$\begin{aligned} \frac{d\psi_1}{d\xi} &\rightarrow -\Sigma^{1/2} & \xi &\rightarrow \infty \\ \frac{d\psi_1}{d\xi} &\rightarrow r_m \Sigma^{1/2} & \xi &\rightarrow -\infty \end{aligned} \quad (41)$$

Now applying the solvability condition,

$$\langle \mathcal{L}\psi_1, \vartheta_m \rangle = \left[ \frac{d\psi_1}{d\xi} \vartheta_m - \psi_1 \frac{d\vartheta_m}{d\xi} \right]_{-\infty}^{\infty} + \langle \psi_1, \mathcal{L}\vartheta_m \rangle = \langle V_2, \vartheta_m^3 \rangle \quad (42)$$

Since  $\vartheta_{m,\infty} = 1$ ,  $\vartheta_{m,-\infty} = r_m$ ,  $\vartheta_{\xi,m,\pm\infty} = 0$ ,  $\mathcal{L}\vartheta_m = 0$  and  $\langle V_2, \vartheta_0^3 \rangle = -\langle V_0, \vartheta_0 \rangle \Delta_m''$ , the above solvability condition becomes simplified as

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$$(1 + r_m)\Sigma^{1/2} = \langle V_0, \vartheta_m \rangle \Delta_m'' \quad (43)$$

Further using the identities  $\Sigma = S/\delta^2$  and  $\Delta' = \delta\Delta_m''$ , the growth rate  $S$  is given in terms of  $\Delta'$  as

$$S = \frac{(\delta\Delta_m'')^2 I^2}{(1 + r_m)^2} = \frac{I^2 \Delta'^2}{(1 + r_m)^2} \quad (44)$$

where the integrated factor  $I = \langle V_0, \vartheta_m \rangle$  and  $I = 2.289$  for  $\gamma = 0$  and  $I \rightarrow 1$  as  $\gamma \rightarrow 1$  (Kim, 1997). For  $\gamma = 0$ , the above growth rate is given by  $S = 1.310\Delta'^2$ .

The numerical results for the growth rate  $S$  with  $K = 0$ ,  $\gamma = 0.0$  and  $L = 1$  is shown in Figure 4 along with the asymptotic relationship given in Eq. (44). In the region of small  $\Delta'$  where variation of  $S$  is quadratic with  $\Delta'$ , the asymptotic relationship shows an excellent quantitative agreement. However, as the value of  $\Delta'$  becomes larger, the variation tends to be linear with  $\Delta'$  and the quantitative agreement holds no longer.

*Planar Instability with General Lewis Numbers.* If Lewis numbers differ from unity, numerical solution is required to obtain the growth rate even for planar disturbances. The numerical results for the planar growth

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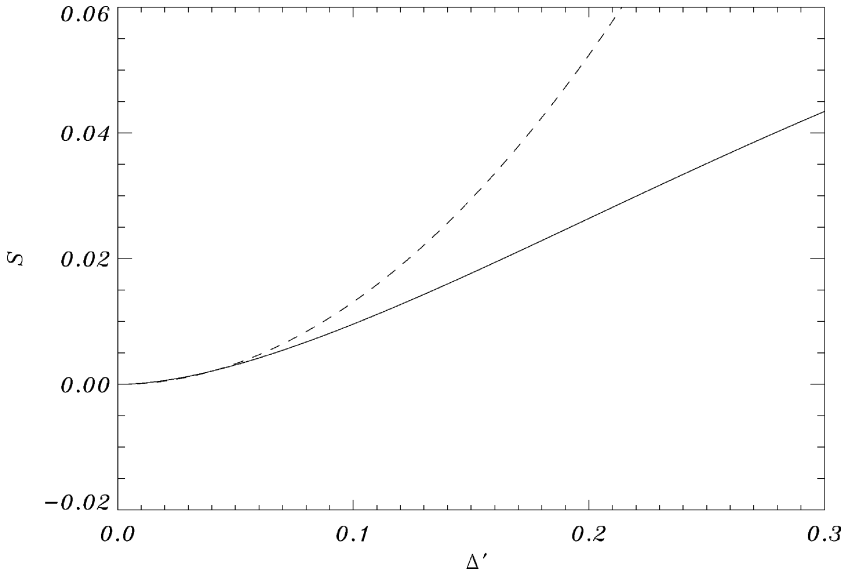


Figure 4. Dependence of  $S(K=0)$  with  $\Delta'$  for  $L=1$  and  $\gamma=0$ .

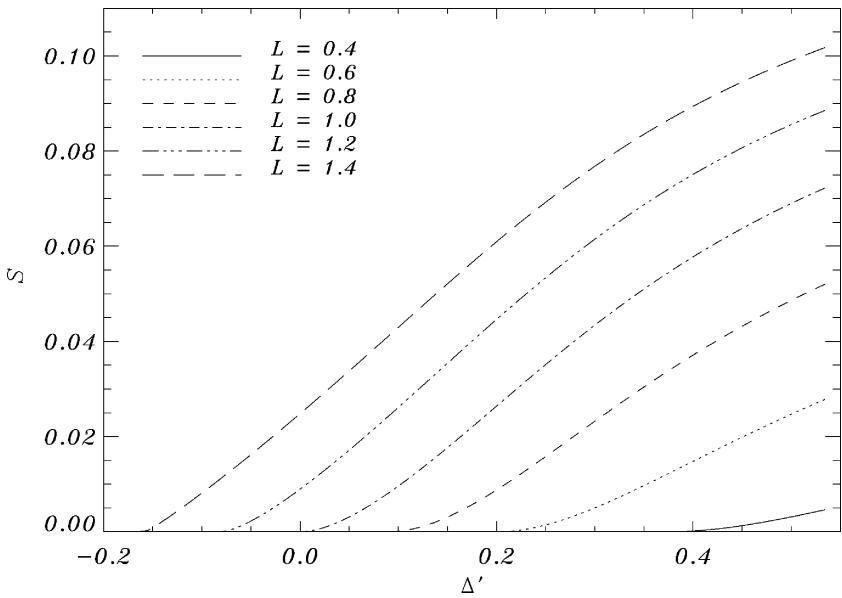


Figure 5. Variation of  $S(K=0)$  with  $\Delta'$  for various values of  $L$  while  $\gamma$  is fixed at zero.

rate  $S$  are shown in Figure 5 for various values of the Lewis number  $L$  as a function of  $\Delta'$ . Perhaps the most outstanding characteristic in Figure 5 is that the onset of the planar growth is delayed to a larger fuel leakage as the Lewis number  $L$  decreases. The numerical values of  $\Delta'$  at the onset condition are compared with Eq. (25). The numerical values are found to be in an agreement with less than 5% error. Since the decrease to the zero growth rate is quadratic, it would be somewhat difficult to pinpoint the exact onset condition. Nevertheless the asymptotic prediction for the onset of the planar instability is found to be quite accurate even for general Lewis numbers.

It is also important to understand the physical meaning of the delayed onset for the planar instability. As the Lewis number decreases below unity by an order of unity, the minimum Damköhler condition given in Eq. (6) no longer serves as the extinction condition. Under this circumstance, the reduced Damköhler number  $\Delta$  has an additional dependence on the fuel leakage to the Damköhler number, which is a true ratio of the characteristic flow time to characteristic chemical time. The additional dependence on the fuel leakage is caused by breaking of the coupling between the energy and species conservation. Consequently, the condition of the minimum Damköhler number does not necessarily correspond to the minimum of  $\Delta$ . In fact, the previous analysis by Kim and Williams (1997) showed that the quasi-steady extinction condition, identified by the minimum Damköhler number, occurs for a fuel leakage much greater than that at the minimum  $\Delta$  condition for  $L < 1$ . The quasi-steady extinction can also be found by the onset of the planar instability for the entire flame structure including the diffusive-convective structure. However, the present analysis does not include the transport in that layer. Nevertheless, the delayed onset of the fast-time planar instability, presented in Eq. (25), gives an approximation to the quasi-steady extinction condition. Even if Eq. (25) does not give an accurate quantitative prediction, it will certainly provide a correct qualitative behavior. In fact, the region between  $\Delta' = 0$  and the delayed onset condition is the window of opportunity, in which the fast-time cellular instability can be realized. Its properties are shown in the following.

### Instabilities for Disturbances with Finite Wave Lengths

As the reduced Damköhler number  $\Delta$  passes the saddle-node bifurcation point, the instability characters are drastically altered. Figure 6 shows the

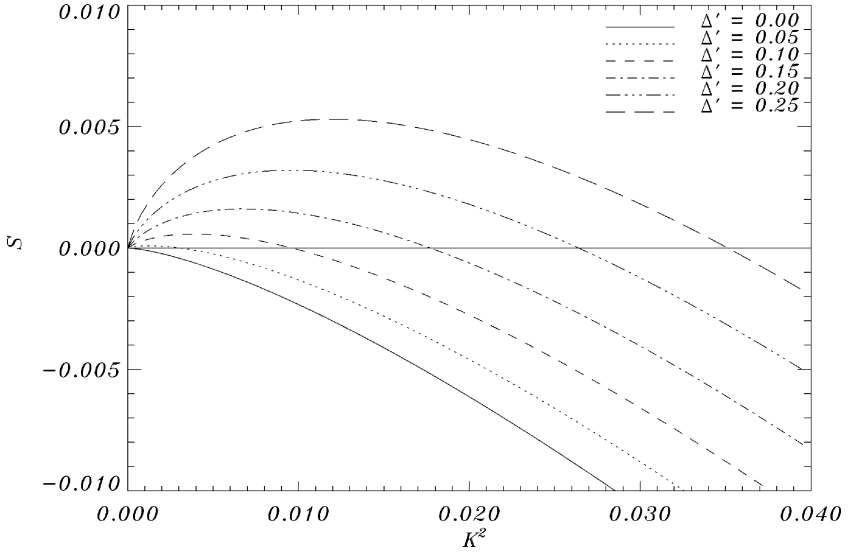


Figure 6. Dispersion relation  $S(K^2)$  for  $\gamma = 0$ ,  $L = 0.4$ , and various values of  $\Delta'$ .

numerical results for the dispersion relation calculated with  $L = 0.4$ . Even if the growth rate for planar disturbances is fixed at zero for  $\Delta' > 0$ , the positive growth rates can now be observed for a finite range of the wave number extending from  $K = 0$ , thereby indicating emergence of the cellular pattern with a relatively large wavelength. In the previous studies on diffusional-thermal instability for diffusion flames, the cellular pattern's wavelength, estimated from the composite solution of the dispersion relationships obtained for the outer convective-diffusive layer and inner diffusive-reactive layer, was found to be quite satisfactory (Kim et al., 1997). Even if the current analysis is carried out with the much shorter inner-zone length scale, the instability is first triggered near the zero wavelength region, so that the results would be applicable to the real cellular patterns found in flames with Lewis numbers much smaller than unity.

It is also worthwhile to note from Figure 6 that the spectral function  $S(K^2)$  looks similar to a parabolic function. An approximate dispersion relationship can be obtained from the asymptotic analysis presented below.

In order to find the approximate dispersion relation, the slope of  $S$  with respect to  $K^2$  at the origin is first considered. Since  $S$  can be

approximated as a linear function of  $K^2$  as  $K \rightarrow 0$ , the asymptotic relation is sought in the form

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$$S = CK^2. \tag{45}$$

Upon substituting this relation into Eq. (4), a set of differential equations is obtained as

$$\left. \begin{aligned} \frac{d^2\psi}{d\xi^2} - (1+C)K^2\psi &= \frac{d^2\chi}{d\xi^2} - (1+LC)K^2\chi = \Delta e^{-(\Theta+\gamma\xi)} [2\Theta\chi - (\Theta^2 - \xi^2)\psi], \\ \frac{d\psi}{d\xi} &\rightarrow \mp \sqrt{(1+C)K^2}\psi, \quad \frac{d\chi}{d\xi} \rightarrow \mp \sqrt{(1+LC)K^2}\chi \quad \text{as } \xi \rightarrow \pm\infty. \end{aligned} \right\} \tag{46}$$

Hereafter we can follow the same procedure that was demonstrated in the previous subsection calculating the onset condition of the planar instability. First  $\sqrt{(1+C)K^2}$  is treated as a small expansion parameter and the resulting equation is compared with Eq. (19). Since the factor  $(1+LC)/(1+C)$  is corresponding to  $L$  in Eq. (19), an asymptotic solution at the marginal stability is found to exist if

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$$\Delta' = \frac{1+r}{2} \left[ 1 - \sqrt{\frac{1+LC}{1+C}} \right]. \tag{47}$$

By solving for  $C$ , the initial slope of  $S$  with respect to  $K^2$  is found to be

$$S = \frac{[2\Delta'/(1+r)][2 - 2\Delta'/(1+r)]}{[1 - 2\Delta'/(1+r)]^2 - L} K^2, \tag{48}$$

The slope is -1 for  $L = 1$  and  $S = 4\Delta'K^2/(1+r_m)(1-L)$  for  $\Delta' \rightarrow 0$ , i.e., near the turning point. Moreover, Eq. (48) clearly shows that the slope becomes positive as soon as  $\Delta'$  becomes positive. Variation of the slope with  $\Delta'$  is shown in Figure 7. Even if there is some quantitative discrepancy for positive values of  $\Delta'$ , the overall agreement is found to be excellent. In particular, the slope diverges as  $\Delta'$  becomes large. This condition can be easily found from the vanishing denominator in Eq. (48) to be  $\Delta' = (1 - \sqrt{L})(1+r)/2$  that is in fact identical to the onset condition for the planar instability. For  $\Delta' > (1 - \sqrt{L})(1+r)/2$ , the linear

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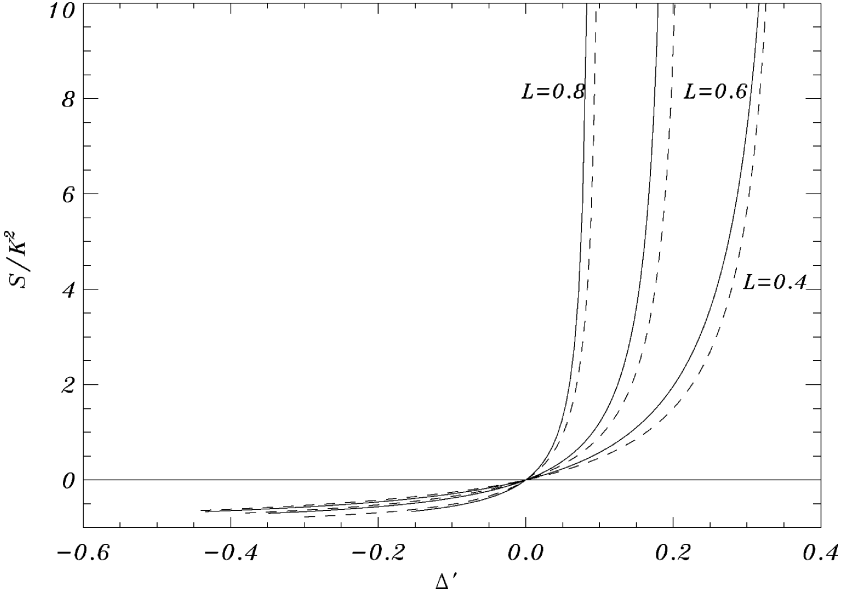


Figure 7. The slope of the curve  $S(K^2)$  at  $K = 0$  for various values of  $L$ . The solid line shows the numerical results. The dashed line is plotted according to Eq. (48).

approximation of  $S(K^2)$  in Eq. (45) is no longer valid since  $S(K = 0)$  takes a positive nonzero value. 605

The spectral behavior for  $\Delta' > (1 - \sqrt{L})(1 + r)/2$  is best shown in Figure 8, in which the growth rate  $S$  is plotted for various values of  $L$  while the values of  $\Delta'$  and  $\gamma$  are fixed at 0.2 and 0.0, respectively. With this parametric condition, the onset Lewis number for the planar instability is found to be 0.64. In Figure 8, the spectral curves for  $L = 0.4$  and  $0.6$  are seen to be anchored at the origin, whereas those for  $L = 1.0$  and  $0.8$  are seen otherwise. Moreover, one must note that all the spectral curves for  $L \leq 1$  return to the same wave number, denoted here by  $K_c$ , when they cross the line of  $S = 0$ . This can be clearly seen from the differential equation shown below, 610

$$\begin{aligned} \frac{d^2\psi}{d\xi^2} - K_c^2\psi &= \frac{d^2\chi}{d\xi^2} - K_c^2\chi = \Delta e^{-(\Theta+\gamma\xi)}[2\Theta\chi - (\Theta^2 - \xi^2)\psi], \\ \frac{d\psi}{d\xi} &\rightarrow \mp\sqrt{K_c^2}\psi, \quad \frac{d\chi}{d\xi} \rightarrow \mp\sqrt{K_c^2}\chi, \quad \text{as } \xi \rightarrow \pm\infty, \end{aligned} \quad (49)$$

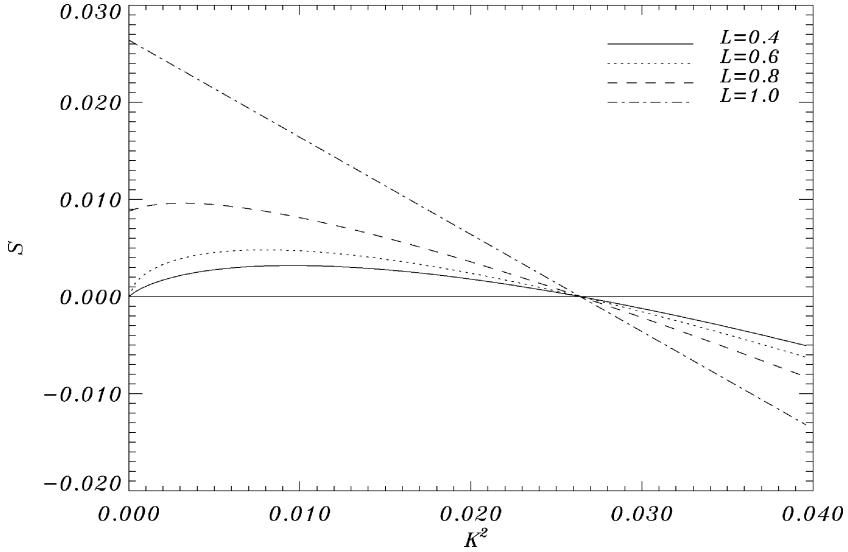


Figure 8. Dispersion relation  $S(K^2)$  for  $\gamma = 0$ ,  $\Delta' = 0.2$ , and various values of  $L$ .

where no dependence on  $L$  is found. The above problem is indeed the problem identical to Eq. (33). Therefore,  $K_c^2$  can be written as

$$K_c^2 = \frac{I^2 \Delta'^2}{(1 + r_m)^2}, \tag{50}$$

where one must bear in mind that the above relationship is valid only near the bifurcation point. In addition, the dispersion relation for  $L = 1$  in the region of small  $\Delta'$  can be written as

$$S + K^2 = \frac{I^2 \Delta'^2}{(1 + r_m)^2} \tag{51}$$

Using Eqs. (44) and (50), the approximate dispersion relation is expressed as

$$S = \frac{4\Delta' K^2}{(1+r)(1-L)} \left(1 - \frac{K^2}{K_c^2}\right). \tag{52}$$

with the wave number for the fastest growing mode corresponding to  $K_c/\sqrt{2}$ .

Based on the above approximate dispersion relation, a nonlinear evolution equation can be guessed. For the distinguished limit of small  $\Delta'$ , the wave number  $K$  is of order  $\Delta'$  and the growth rate  $S$  is of order  $\Delta'^3$ . The nonlinear term which eventually limits the amplitude of the growing perturbation can be postulated to be the simple cubic term since the inner structure of diffusion flame does not possess any propagating nature. The time coordinate  $t$ , transverse coordinate  $z$ , and perturbation  $\psi$  are rescaled by  $\Delta'^{-3}$ ,  $\Delta'^{-1}$  and  $\Delta'^{-1}$ , respectively. Then, the nonlinear evolution equation would take a form of

$$\Psi_\tau + \Psi_{\zeta\zeta} + \Psi_{\zeta\zeta\zeta} + \Psi^3 = 0 \quad (53)$$

where  $\tau$ ,  $\zeta$ , and  $\Psi$  are the rescaled stretched variables with a set of proper conversion factors to adjust the coefficient to each term at the above nonlinear equation.

The above nonlinear equation resembles the Kuramoto-Sivhashinsky equation for its linear terms and the Landau-Ginzburg equation for its nonlinear term. Since the basic nature of nonlinear evolution is more strongly dependent on the nonlinear term, we would anticipate that the corresponding nonlinear evolution will be similar to that of the Rayleigh-Benárd instability, which is described by the Landau-Ginzburg equation. Such nonlinear character is already seen in the numerical simulation of cellular patterns in diffusion flames (Lee and Kim, 2000). In their numerical simulation, the initial growth was quite similar to that of premixed flames as guessed by the similar linear terms; however, the nonlinear term eventually takes over the final stage of nonlinear evolution to form a series of stationary stripes as seen in the Rayleigh-Benárd instability.

### Comparison with the Conventional Diffusional-Thermal Instability

The results of the present fast-time instability exhibit two distinct differences from the instability characteristics obtained by the linear stability analysis employing the NEF limit (Kim, 1997). First, the present fast-time instability shows that the growth rate is zero at the origin while the diffusional-thermal instability shows negative growth rate before reaching the extinction condition. This is perhaps caused by ignoring the outer convective-diffusive layer where the damping effect for long wave disturbances exists. The diffusion effect in the inner layer is not felt

by the long wave disturbances and the stability analysis using the outer-layer scaling would be necessary to achieve proper damping effect.

The lack of long wave damping effect appears in the range of Lewis number for the cellular instability too. The present analysis predicts the cellular instability for the entire Lewis number range below unity. However, the diffusional-thermal instability predicts a small range of the Lewis number at  $O(1/\beta)$  below unity Lewis number, not exhibiting any cellular instability. In this parametric range of the Lewis number, in view of Eq. (32), the meaningful  $\Delta'$  is found to be of  $O(1/\beta)$  and correspondingly  $K_c$  is also found from Eq. (50) to be of  $O(1/\beta)$  that is the same order of magnitude for the wave number in the diffusional-thermal instability. Since the fast-time instability does not possess the damping effect for such small long wave, the present analysis does not show any region of the Lewis number below unity without cellular instability. However, one must keep in mind that the Lewis number range without cellular instability is asymptotically small as  $\beta \rightarrow \infty$ . Thus the critical Lewis number of  $L = 1$  is indeed correct from the viewpoint of order unity scaling. In addition, for sufficiently small Lewis numbers, the present analysis should be quite capable of predicting the proper instability characters.

## CONCLUDING REMARKS AND FUTURE WORKS

The fast-time instability of diffusion flames is studied as a model problem for the cellular instability in diffusion flames with Lewis numbers far from unity by an amount of order unity. Particular attention is focused on the stability of the inner reactive-diffusive zone structure of near-extinction diffusion flames. Consequently, the present study is devoted to examining the stability of the Liñán's diffusion-flame regime with deviation of the Lewis number from unity by an amount of order unity while employing the fast inner-zone time and length scales.

The stability analysis is carried out asymptotically and numerically to exhibit the full spectral nature, particularly in the neighborhood of the saddle-node bifurcation condition corresponding to the minimum reduced Damköhler number  $\Delta$  that is commonly known as the Liñán's diffusion-flame extinction condition. The present analysis explains that the minimum  $\Delta$  condition does not necessarily correspond to the extinction condition, and flames with Lewis numbers less than unity can survive beyond the minimum  $\Delta$  condition. Furthermore, the cellular instability could begin to emerge upon passing this saddle-node bifurcation

condition. It is thus predicted that the cellular instability is observable only for near-extinction diffusion flames with Lewis numbers less than unity, as previously predicted by the experimental studies as well as by the linear stability analysis employing the NEF limit. However, it must be emphasized again that the present results obtained by calculating the fast-time instability is valid only for  $(1 - L) = O(1)$ . Under the fast-time scaling employed for this distinguished limit, the damping effect arising from the convective-diffusive layer does not come into play, so that the damping of long wave disturbances and the stable region of Lewis number just below unity are not predicted in this fast-time instability. However, one must keep in mind that in the current distinguished limit these effects lie in the Lewis number's inner layer located around the unity Lewis number, so that the leading-order behavior is not contradictory with the previous experimental and analytical results.

The present study can be further extended to the different fast-time instability problems. Immediately, the analysis can be extended to the case of Lewis number greater unity, where oscillatory instabilities are anticipated. For this analysis, care must be taken when the numerical analysis is carried out because the eigenfunction will oscillate while converging to the boundary condition and the imposition of the boundary condition could be a tricky matter. Because of this complication, the Evans function technique is expected to play a key role in calculating the eigensolutions. Another interesting distinguished limit corresponds to the limit, in which the heat loss factor  $\gamma$  in Eq. (1) approaches unity, formally known as the Liñán's premixed flame regime. Under this distinguished limit, the factor  $(1 - \gamma)$  can be used as an expansion parameter to scale the minimum  $\Delta$  condition and the reactant leakage  $\alpha$ . Because the previous analyses predicted that the instability character differs across the adiabatic condition, the spectral characteristics will be examined to better understand how the stability of the inner-layer structure is characterized for near-adiabatic diffusion flames.

## APPENDIX: DERIVATION OF THE MINIMUM $\Delta$ CONDITION

If the parameter  $\gamma$  approaches  $\pm 1$  (i.e., the flame is nearly adiabatic), Eq. (1) exhibits an asymptotic solution, in which  $m \equiv (1 - |\gamma|)/2$  is adopted as the smaller expansion parameter. Since the cases of positive and negative  $\gamma$  are symmetric, here we only consider the case of  $\gamma \rightarrow -1$ , the case arising for large oxidizer leakage. This convention is used to formulate the problem

in a much more familiar flame configuration in which the premixture is approaching from the negative infinity. The procedure shown here follows a previous analysis by Clavin and Liñán (1984). 740

To proceed with the analysis, it is convenient to introduce new variables as

$$\eta = 2\xi - \alpha^- = 2\xi - \frac{a}{m} \quad \varphi = \Theta - \xi \quad (\text{A.1})$$

where an alternative heat-loss parameter  $m$  is defined as

$$m \equiv \frac{1 - |\gamma|}{2} \quad (\text{A.2})$$

and the oxygen-leakage parameter  $\alpha_O = \alpha^- = \alpha(-\gamma)$  is rescaled according to  $\alpha^- = a/m$  with  $a$  being of order unity. For small values of  $m$ , we pose the problem that of finding the reduced Damköhler number  $\Delta$  that corresponds to a given value of the rescaled fuel-leakage parameter  $a$ . Upon substitution of the new variables in Eq. (A.1) into Eq. (1), the inner equation becomes 750

$$\begin{aligned} \frac{d^2\varphi}{d\eta^2} &= \Delta \frac{ae^{-a}}{4m} \varphi \exp[-(\varphi + m\eta)] \left[ 1 + \frac{m}{a}(\varphi + \eta) \right] \\ \varphi_\infty &\rightarrow 0 \quad (\varphi + \eta)_{-\infty} \rightarrow 0 \end{aligned} \quad (\text{A.3})$$

With the parameter  $m$  taken as the small expansion parameter, the solution is sought in the form 755

$$\left. \begin{aligned} \varphi &= \varphi_0 + m\varphi_1 + \dots \\ \delta &= \Delta \frac{ae^{-a}}{4m} = \delta_0(1 + md + \dots) \end{aligned} \right\} \quad (\text{A.4})$$

where  $\varphi_0$ ,  $\varphi_1$ , as well as  $\delta_0$  and  $d$  are assumed to be of order unity.

At the leading order, we find

$$\begin{aligned} \frac{d^2\varphi_0}{d\eta^2} &= \delta_0\varphi_0 \exp(-\varphi_0) \\ \varphi_0 &\rightarrow 0 \quad \text{as} \quad \eta \rightarrow \infty \quad \varphi_0 + \eta \rightarrow 0 \quad \text{as} \quad \eta \rightarrow -\infty \end{aligned} \quad (\text{A.5})$$

The energy integral of the above equation results in  $\delta_0 = 1/2$ , thereby providing the first approximation to  $\Delta$ ,

$$\Delta = \frac{2m}{a} \exp(a) = \frac{2}{\alpha^-} \exp(m\alpha^-) \quad (\text{A.6})$$

The equation for  $\varphi_1$  of the expansion in Eq. (A.3) is

$$\frac{d^2\varphi_1}{d\eta^2} - \delta_0(1 - \varphi_0) \exp(-\varphi_0)\varphi_1 = \delta_0\varphi_0 \exp(-\varphi_0) \left[ d - \eta + \frac{\varphi_0 + \eta}{a} \right] \varphi_1 \rightarrow 0 \text{ as } \eta \rightarrow \pm\infty \quad (\text{A.7})$$

The above differential equations can be written in a more standard form

$$\mathcal{L}(\varphi_1) = \delta_0 \varphi_0 \exp(-\varphi_0) \left[ d - \eta + \frac{\varphi_0 + \eta}{a} \right] \varphi_1 \quad (\text{A.8})$$

where the linear operator  $\mathcal{L}$  is defined to be

$$\mathcal{L} = \frac{d^2}{d\eta^2} - \delta_0(1 - \varphi_0) \exp(-\varphi_0)$$

The homogeneous solution  $\varphi_h$  to the linear operator  $\mathcal{L}$  is simply found to be

$$\varphi_h = \frac{d\varphi_0}{d\eta} \quad (\text{A.9})$$

Then, the solution to Eq. (A.7) exists if its project to  $\varphi_h$  vanishes. From the solvability condition, we find an algebraic identity

$$\int_{-\infty}^{\infty} \delta_0 \varphi_0 \exp(-\varphi_0) \left[ d - \eta + \frac{\varphi_0 + \eta}{a} \right] \frac{d\varphi_0}{d\eta} d\eta = 0 \quad (\text{A.10})$$

The integration yields

$$-d - v + \frac{v - 2}{a} = 0 \quad \implies \quad d = -v - \frac{2 - v}{a} \quad (\text{A.11})$$

where use has been made of the integral identities

$$\int_{-\infty}^{\infty} \varphi_0 \exp(-\varphi_0) \frac{d\varphi_0}{d\eta} d\eta = \int_0^{\infty} \varphi_0 \exp(-\varphi_0) d\varphi_0 = -1$$

$$\int_{-\infty}^{\infty} \varphi_0^2 \exp(-\varphi_0) \frac{d\varphi_0}{d\eta} d\eta = \int_0^{\infty} \varphi_0^2 \exp(-\varphi_0) d\varphi_0 = -2$$

and the constant  $v$  is

$$v = \int_{-\infty}^{\infty} \varphi_0 \exp(-\varphi_0) \frac{d\varphi_0}{d\eta} \eta d\eta = 1.3440$$

which is obtained by a numerical integration. Consequently, the two-term expansion for  $\delta$  is found to be

$$\delta = \Delta \frac{ae^{-a}}{4m} = \frac{1}{2}(1 - 1.3440m - 0.6560m/a + o(m)) \quad (\text{A.12})$$

Finding the root for the derivative of the logarithm for the above equation yields the two-expansion for the condition for minimum  $\Delta$  such as 785

$$a = 1 - 0.6560m = 1 - (2 - \nu)m \quad (\text{A.13})$$

and the corresponding two-term expansion for  $\delta_E$  or  $\Delta_E$  is

$$\delta_E = 2em(1 - 2m) \quad \text{or} \quad \Delta_E = e\{(1 - |\gamma|) - (1 - |\gamma|)^2\} \quad (\text{A.14})$$

Of course, the above expression is the first two terms of the so-called 790 Liñán's diffusion-flame extinction criterion. Moreover, from a numerical calculation, one easily finds that the minimum  $\Delta$  for  $\gamma = 0$  is  $\Delta_E(\gamma = 0) = 0.315e = 0.856$ . Imposing the above condition and the symmetry of  $\Delta_E$  at  $\gamma = 0$ , the four-term expansion of  $\Delta_E$  becomes

$$\Delta_E = e\{(1 - |\gamma|) - (1 - |\gamma|)^2 + 0.26(1 - |\gamma|)^3 + 0.055(1 - |\gamma|)^4\} \quad (\text{A.15})$$

The above extinction criterion is found to be in excellent agreement with the numerical results for  $\Delta_E$  throughout the entire range of  $\gamma$ , i.e.,  $-1 < \gamma < 1$ .

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