

Theory & modeling of detonation wave stability: A brief look at the past and toward the future.†

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1. Introduction

A detonation is a form of propagating wave front that can occur in gaseous, liquid or solid explosives. It has a structure consisting of a lead shock which is sustained by chemical reaction in the flow downstream of the shock. In gaseous mixtures, detonation fronts tend to propagate in a highly unsteady manner, the most common illustration of which is the formation of spectacular fish-scale patterns on the walls of rectangular shock tubes lined with soot-covered aluminum foil (Fickett & Davis 1979). On the other hand, in condensed-phase explosives there is no definitive experimental evidence as to the fine-scale structure of propagating detonation fronts, and thus questions pertaining to their stability properties remain unanswered at present. Understanding and modeling the dynamics of the evolution of unstable detonation fronts is an essential part of any application that utilizes the rapid energy conversion rates of detonation waves for practical purposes, such as pulsed detonation engines for propulsion applications or precision directed energy release through shaped detonations in condensed phase explosives.

The purpose of this talk is to give an overview of our current state of knowledge in the area of detonation wave stability theory. This abstract is a shortened and modified version of an extended review paper currently being written by the author in the area of detonation stability. In particular, due to space and time limitations, here we will concentrate on a summary of the results of a number of papers that have appeared since 1990, following a paper by Lee & Stewart (1990) on the normal mode analysis of the linear stability of planar ZND detonations that stimulated a renewed interest in the area. The list of results mentioned below is certainly not exhaustive, and no offence is intended by the omission of any one result. A more complete review will appear in the future.

In the following, I will briefly review results from: formal linear stability theory for gas phase detonations with simple and multi-step reaction kinetics; asymptotic studies on the linear and nonlinear evolution of unstable detonation fronts based on various limiting cases (including slowly evolving, weakly curved approximations, as well as high-overdrive and weak heat release limits); and relationships between predictions of the linear stability analysis and direct numerical simulation of detonation wave evolution for pulsating and cellular instabilities. Finally, I will highlight some future challenges for detonation wave stability theory, including the need for different asymptotic approaches for resolving the practical issue of the evolution of Chapman-Jouguet detonations, analyses of detonation stability for full reaction chemistry, and the study of detonation in condensed-phase (high-explosive) systems.

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2. Steady planar detonation wave properties

Before discussing the stability analysis of planar detonation waves, we need to review the basic properties of the steady planar detonation structure wave. An extensive review is given in Fickett & Davis (1979). The mathematical and numerical modelling of detonation waves is almost invariably undertaken in the context of the compressible, inviscid Euler equations of motion accounting for heat addition from chemical reaction. All thermodynamic variables are assumed to be in local thermodynamic equilibrium. Steady, planar detonation wave solutions can be constructed in the context of this model. The most familiar are known as ZND waves, after Zeldovich, von-Neumann and Döring (Fickett & Davis 1979). The ZND wave profile consists of a sharp (discontinuous) shock front, followed by a finite zone of chemical reaction, in which the flow is subsonic relative to the shock, which terminates at a location known as the “final point”. The (inert) flow downstream of the final point is dependent on the nature of the rear boundary. Fickett & Davis (1979) restrict the term ZND to the case of a single or sequence of reactions of positive thermicity (a quantity which measures the conversion of chemical bond energy to macroscopic translational energy), and we will adopt this convention in the following. Each wave of ZND type has a minimum sustainable detonation speed, D_{min} , known as the Chapman-Jouguet (or CJ) speed in which a sonic flow point (relative to the detonation shock speed) arises at the final point. Such waves have the property that the reaction zone between the shock and sonic point is acoustically isolated from the flow structure downstream of the sonic point. Importantly, the speed of, and the thermodynamic state at the final point in, the CJ wave are determined by the initial upstream shock state and by the equation of state for the reaction products alone. The particular form of the reaction rate law and the equation of state for partially reacted material affect only the interior structure of the reaction zone. Any ZND wave travelling with a speed higher than D_{min} is called overdriven, and must be mechanically supported in some fashion, e.g by a driving piston. In this case, the region of flow between the shock and supporting boundary is subsonic, and between the final point and the supporting boundary there is a region of spatially-uniform flow. For waves of CJ type, the detonation can be supported, in which case the region downstream of the sonic point consists of spatially uniform flow, or underdriven, in which case there is region of unsteady supersonic flow downstream of the sonic point, where an expansion (or Taylor) wave connects the state at the final point to the rear boundary. The Taylor wave may or may not be attached to the final point.

More complicated steady, planar detonation wave solutions can be constructed if the restrictions that lead to the ZND wave are lifted. Typically, these arise for more complex chemistry models, e.g. two- or three-step models that better mimic the reaction structure profiles of real detonations. If each of the reaction steps is taken to be irreversible, and such that the thermicity function remains positive throughout the reaction zone, then the solution properties are similar to those of the detonation wave profiles described above. However, an important class of steady solution can also occur with more complex chemistry models, and these arise when the sonic point appears at a point of incomplete reaction. In order for this to happen, the chemistry must be such that the thermicity can vanish at the sonic point. Such solutions are called weak, pathological or eigenvalue waves (Fickett & Davis 1979). They can occur, for example, in systems with a single irreversible reaction in which a mole decrement arises in the transition from fuel to product, or for a two-step sequential reaction in which the second reaction is endothermic. Such solutions again represent the minimum propagation speed of a steady planar wave under that particular form of chemistry. In this case, though, the detonation speed and the state at the sonic point do now depend on the reaction rate model and the equation of state for

partially reacted material. Between the internal sonic point and the final point, the flow may be supersonic, with the final point lying on a weak point of the product Hugoniot curve, or subsonic, with the final point lying on a strong point of the product Hugoniot curve.

These steady propagating wave structures are the starting point for a number of stability investigations that have been performed since 1990 that are described below.

3. Linear Stability analysis: Normal-mode analysis

The first issue we review is that of the formal linear stability analysis of steady detonation waves without any asymptotic approximation of the underlying steady state. An extensive study of this problem was undertaken by Jerry Erpenbeck in the 1960s using an initial value Laplace transform approach (e.g. Erpenbeck 1964), and an excellent summary of Erpenbeck's results, and others, is given in Fickett & Davis (1979). In 1990, Lee & Stewart published the results of a formal normal-mode approach to the problem of linear detonation stability for a one-step Arrhenius reaction model with simple (unit reaction order) reactant depletion. Below we review the normal-mode stability analysis applicable to steady detonation waves governed by an arbitrary caloric equation of state. Although the specification for an arbitrary number of reactant species and reaction steps is simple, due to space limitations we will restrict our attention here to the case of a single step reaction with an arbitrary rate law.

3.1. Equations

The nondimensional Euler equations of motion coupled with an equation for species conservation for the single-step reaction $\mathcal{F} \rightarrow \mathcal{P}$ are given by

$$\frac{D\Lambda}{Dt} - \Lambda (\nabla^l \cdot \mathbf{u}^l) = 0, \quad \frac{D\mathbf{u}^l}{Dt} = -\Lambda \nabla^l p, \quad \frac{De}{Dt} = -p\Lambda (\nabla^l \cdot \mathbf{u}^l), \quad \frac{D\beta}{Dt} = r, \quad (3.1)$$

for specific volume Λ , pressure p , specific internal energy e , laboratory frame velocity $\mathbf{u}^l = (u^l, v^l)$ and reaction progress variable β for the single-step reaction, where $\beta = 1$ represents unreacted fuel, and $\beta = 0$ is fully depleted fuel. At this stage we adopt the general caloric equation of state and reaction rate forms,

$$e = e(p, \Lambda, \beta), \quad r = r(p, \Lambda, \beta). \quad (3.2a,b)$$

The (chemically) frozen sound speed c is related to (3.2a) via

$$c^2 = \Lambda^2 (p + e_{,\Lambda}) / e_{,p}. \quad (3.3)$$

Equations (3.1)–(3.3) have been nondimensionalized such that $\Lambda = \tilde{\Lambda} / \tilde{\Lambda}_0$, $\mathbf{u}^l = \tilde{\mathbf{u}}^l / \tilde{D}$, $p = \tilde{\Lambda}_0 \tilde{p} / \tilde{D}^2$, $\mathbf{x}^l = \tilde{\mathbf{x}}^l / \tilde{l}$, $t = \tilde{D} \tilde{t} / \tilde{l}$, $e = \tilde{e} / \tilde{D}^2$, $c^2 = \tilde{c}^2 / \tilde{D}^2$, where $\tilde{\Lambda}_u$ is the upstream (ambient) specific volume and \tilde{D} is the dimensional planar steady detonation velocity. The length scale \tilde{l} is set by the particular rate form under study.

3.2. Travelling wave solutions

For thermodynamically consistent forms of (3.2a), the above model supports a one-dimensional steady, travelling wave solution consisting of a lead shock followed by a region of chemical reaction. In a reference frame attached to the wave, $x = x^l - t$, where $x = 0$ is set by the location of the shock front, the thermodynamic states in the steady wave for a general caloric equation of state are connected by the conditions

$$\Lambda = -u, \quad p = u + 1 + p_0, \quad e + p\Lambda + u^2/2 = e_0 + p_0 + 1/2, \quad (3.4a,b,c)$$

where $p_0 = \tilde{\Lambda}_0 \tilde{p}_0 / \tilde{D}^2$ (\tilde{p}_0 is the dimensional unshocked material pressure), $e_0 = \tilde{e}_0 / \tilde{D}^2$ is the ambient internal energy, and $u = u^l - 1$. Relations (3.4a,b) define the Rayleigh line variation, which holds regardless of the form of (3.2a), while (3.4c) defines the Hugoniot curves for any degree of reaction, which do depend on the form of (3.2a). The spatial structure of the ZND wave can be determined through the Master equation relation,

$$u_{,x} = -\frac{ue_{,\beta}}{\eta e_{,p}} r, \quad \eta = u^2 - c^2, \quad (3.5)$$

which depends on the form of both e and r . Here, η is a sonic parameter. The structure of the CJ wave, the slowest of all possible steady wave solutions, is defined by the appearance of a sonic point relative to the detonation wave speed either at a point of incomplete or complete reaction. For general forms of (3.2a,b) this may be determined as follows. If \tilde{D}_{CJ} , \tilde{p}_0 and \tilde{e}_0 are known initially, where \tilde{D}_{CJ} is the CJ speed, (3.4a,b,c) can be solved to determine the immediate post-shock state at which no reaction has occurred. Subsequently, (3.5) can be integrated from the shock ($x = 0$) into the region $x < 0$. A single value of the heat of reaction \tilde{q} will define a solution trajectory which passes through the critical point (where η and r vanish simultaneously), and it is this trajectory which defines the spatial structure of the CJ wave. Note that if the sonic point appears at the end of the reaction zone, then (3.4a,b,c) can be used to determine \tilde{q} algebraically, otherwise \tilde{q} must be determined iteratively by successive integrations of (3.5). If \tilde{q} , \tilde{p}_0 and \tilde{e}_0 are known initially, a similar procedure can be used to determine \tilde{D}_{CJ} . Finally, once the CJ wave structure has been determined, it is straightforward to generate the spatial structure of the overdriven wave, defined by a given overdrive factor $f = \tilde{D}^2 / \tilde{D}_{CJ}^2$.

3.3. Perturbation equations

The equations governing small (linear) perturbations to the steady traveling wave identified in §3.2 are constructed as follows. We transform to a new spatial coordinate system $x = x^l - t - \Psi(y, t)$, $y^l = y$, where $x^l = t + \Psi(y^l, t)$ is the shock locus in laboratory frame, which now becomes $x = 0$. We seek a normal mode decomposition,

$$\Psi = \Psi_0 \exp(\alpha t + iky), \quad \mathbf{z} = \mathbf{z}^* + \Psi_0 \mathbf{z}'(x) \exp(\alpha t + iky), \quad (3.6)$$

for the growth rate/frequency eigenvalue α and wavenumber k , where $\mathbf{z} = (\Lambda, u, v, p, \beta)^T$ represents the vector of dependent variables, the superscript $*$ refers to the underlying steady wave solution, the $'$ quantities indicate the spatially (x) dependent eigenfunctions and $\Psi_0 \ll 1$. The system of equations that govern the linear stability of a detonation with the general caloric equation of state and reaction rate law (3.2a,b) is then determined to be

$$\eta^* u^* \mathbf{z}'_{,x} + \mathbf{A}^* \mathbf{z}' + \mathbf{a}^* = 0, \quad (3.7)$$

3.5.1. Overdriven waves

The closure condition for supported overdriven waves of finite length assumes that the amplitude of the signal on all the characteristics in the equilibrium (fully reacted) zone that point toward the reaction zone is zero (Short *et al.* 2005). For unstable modes, this also ensures that the eigenfunction corresponding to forward wave propagation at $x = \infty$ is spatially bounded. For the arbitrary equation of state (3.2a), this condition becomes

$$u' - \frac{u_b \omega}{c_b} p' - \frac{ik u_b}{\alpha} v' = 0, \quad \omega = \sqrt{1 - \eta_b k^2 / \alpha^2}. \quad (3.14)$$

where the subscript b denotes steady conditions in the equilibrium zone. For reaction rates that lead to steady detonation wave profiles formally having infinite spatial extent, condition (3.14) also ensures bounded eigenfunction solutions.

3.6. CJ waves

Steady, traveling Chapman-Jouguet waves possess a bounding, forward facing sonic characteristic (where $\eta = 0$), with the implication that when (3.7) is integrated for $x < 0$ for arbitrary α , using (3.12) at $x = 0$, \mathbf{z}' will contain a single spatially unbounded mode for $\text{Re}(\alpha) \geq 0$ at the steady sonic point. Elimination of the unbounded mode leads to the closure condition

$$p' + u' - ik\alpha^{-1} u v' + 2u_{,x} \alpha^{-1} (u' - \alpha) + b_p p' + b_\Lambda \Lambda' + b_\beta \beta' = 0 \quad (3.15)$$

where b_p , b_Λ and b_β are constants that are determined in terms of the steady state conditions at the sonic flow point (Short, Bdzil & Anguelova 2005). Equation (3.15) is applied at the point in the steady wave where $u = -c$. It again applies for general forms of (3.2a,b), and also regardless of whether $\eta = 0$ occurs at a point of incomplete reaction or at the point of complete reaction. Also, depending of the form of (3.2a,b) not all the terms in (3.15) will be important. Note that there are certain situations where (3.15) will not ensure bounded solutions, and these are related in one case to specific singular forms of the reaction rate (Short *et al.* 2005). It is important to note that in general one has no ability to impose flow conditions along sonic characteristics, and thus it is formally not correct to apply (3.14) in the limit $u \rightarrow -c$ as the closure condition for CJ waves. In many cases, though, (3.14) in the limit $u \rightarrow -c$ and (3.15) do reduce to the same condition.

3.7. Calculation of α and $\mathbf{z}'(x)$

The main (numerical) strategy that has been used to evaluate the spectrum for α and $\mathbf{z}'(x)$ is as follows. The system (3.7) is integrated from $x = 0$ using (3.12) into the region $x < 0$. An eigensolution is obtained when (3.14) is satisfied at the equilibrium point for overdriven waves, and (3.15) is satisfied arbitrarily close to the sonic point for CJ waves. Note that there are some interesting consequences of this strategy in which an eigenvalue α can be calculated correctly, but its corresponding eigenfunction is in error (Short *et al.* 2005). This calculation procedure has been used to obtain all the results described below, except those described in §4.3, where an alternatively reverse integration strategy developed by Sharpe (1999) was used.

4. Linear stability: Gaseous detonations and dependence on rate models

Before discussing particular results, we note that for historical reasons the analysis of detonation stability for individual rate models has been conducted for a variety of

nondimensional scalings based on the underlying steady-state wave structure. There are three main scalings in use, which I will denote for the purposes of this review as the Erpenbeck scalings (nondimensionalisation based on ambient upstream conditions), Bdzil scalings (based on ZND detonation velocity scalings) and those based on ZND post-shock conditions. Unfortunately, the different flavors of nondimensionalisation are often a source of confusion. However, each scaling has its own advantages, and I will adhere in the following to the historical scalings used for studying each type of the reaction rate model. Conversions between the different scalings are straightforward. Also, for each of the reaction rate models discussed below, the ideal gas equations of state

$$e = \frac{p}{(\gamma - 1)\rho} - q, \quad T = \frac{\gamma p}{\rho} \quad (4.1)$$

are used where q is the heat release function and the adiabatic gamma is assumed to be constant.

4.1. One-step irreversible Arrhenius models

Much of the analysis of the linear stability of detonation has been conducted for one-step irreversible Arrhenius models, i.e. fuel (F) \rightarrow product (P) via a thermal decomposition reaction, where

$$r = k(1 - \lambda)^\nu \exp(-E/T), \quad q = Q(1 - \lambda) \quad (4.2)$$

where E and Q are the Erpenbeck scaled activation energy and heat release, ν is the reaction order and $\lambda (= 1 - \beta)$ is the mass fraction of fuel. The normal mode analysis of the linear stability of planar ZND detonations, without invoking any asymptotic approximations, has been extensively studied for the one-step reaction (e.g. Lee & Stewart 1990, Bourlioux & Majda 1992, He & Lee 1995, Sharpe 1997, Short 1997, Short & Stewart 1998, Short & Wang 2001). There are five main bifurcation parameters in the problem; the detonation overdrive f , the activation energy E , the heat release Q , the ratio of specific heats γ , and the reaction order ν . Variations in any of these quantities affect the domains between instability and stability of the planar ZND detonations. These changes may be understood in the context of the variation in the underlying ZND profile that each change in a bifurcation parameter invokes. This is discussed in detail in Short & Stewart (1998), and can be related to the change in the rate of heat release in the ZND profile; the more rapid the heat release the greater tendency for instability. Figure 1 illustrates some of the neutral stability boundary variations that have been calculated for the one-step reaction. Although there are exceptions to these general trends, basically increasing overdrive, decreasing activation energy, decreasing heat release and increasing reaction order tend to stabilize the detonation.

Within the context of the one-step reaction, other studies have considered the normal mode linear stability of quasi-steady, weakly-curved detonations (Watt & Sharpe 2004), spinning detonations (Kasimov & Stewart 2004) and detonations with friction losses (Dionne *et al.* 2000).

4.2. Chain-branching type reaction models

One of the drawbacks of the one-step Arrhenius reaction model is its inability to reproduce separate finite regions of induction and main heat release layers, a feature of many detonations where fuel is converted into product via a series of chain-branching type reactions. For example, for high activation energies, an induction zone can be reproduced with the one-step reaction, but the main reaction layer is then spatially thin relative to the induction layer (this is the limiting square wave detonation structure). To ascertain the effect that chain-branching type reactions have on detonation stability, Short & Dold

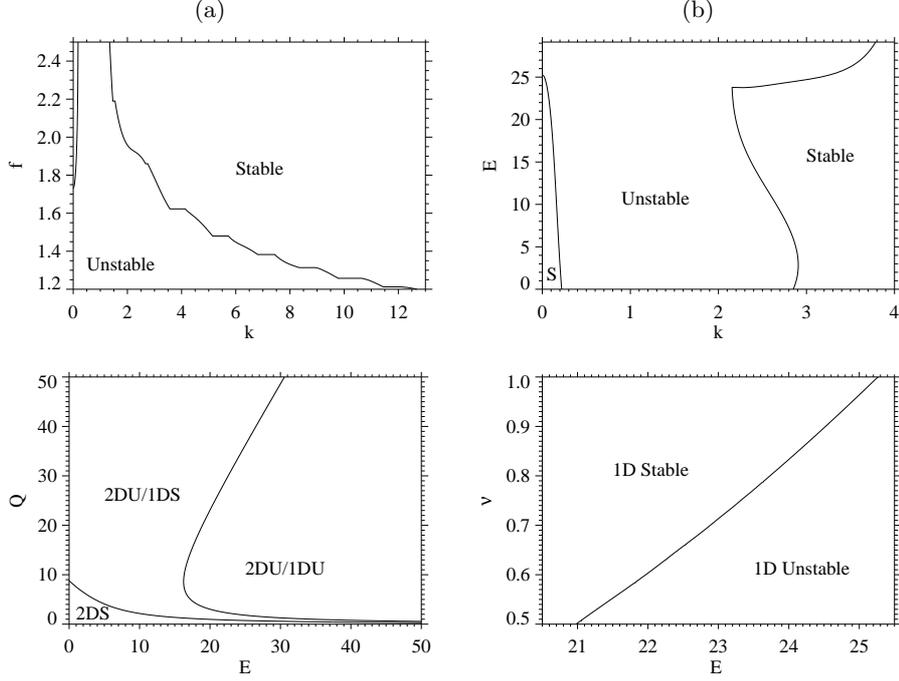
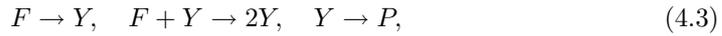


FIGURE 1. Neutral stability boundaries for the one-step Arrhenius reaction rate. (a) 2D neutral stability boundary in overdrive-wavenumber space, for $Q = 50$, $E = 50$, $\gamma = 1.2$ and $\nu = 1$. (b) 2D boundary in activation energy against wavenumber space for $Q = 50$, $f = 1$, $\gamma = 1.2$ and $\nu = 1$. (c) 1D and 2D $Q - E$ neutral stability boundaries for $f = 1.2$, $\gamma = 1.2$ and $\nu = 1$. (d) 1D neutral stability boundary in reaction order-activation energy space for $Q = 50$, $f = 1$ and $\gamma = 1.2$.

(1996) and Short & Quirk (1997) considered a normal mode analysis of detonation stability for a model three-step chain-branching that can reproduce the essential features of more complex chain-branching reaction mechanisms. The three step mechanism is given by



for fuel F , radical species Y and product P . The first is the initiation reaction, the second the chain-branching reaction and the third the chain-termination reaction, with rates r_I , r_B and r_C respectively. The initiation and branching reactions are assumed to have a temperature-dependent Arrhenius form, while the termination reaction has a fixed rate, independent of temperature. Non-dimensional consumption equations for fuel (f) and radical (y) mass fractions are

$$\frac{Df}{Dt} = -r_I - r_B, \quad \frac{Dy}{Dt} = r_I + r_B - r_C, \quad (4.4)$$

with corresponding reaction rates,

$$r_I = f \exp \left[\theta_I \left(\frac{1}{T_I} - \frac{1}{T} \right) \right], \quad r_B = fy \exp \left[\theta_B \left(\frac{1}{T_B} - \frac{1}{T} \right) \right], \quad r_C = y. \quad (4.5)$$

Here T_I and T_B are the chain-initiation and chain-branching cross-over temperatures respectively, i.e. the temperatures at which the initiation and branching rates are of the same order as the termination rate. The two activation energies are θ_I and θ_B . Conditions

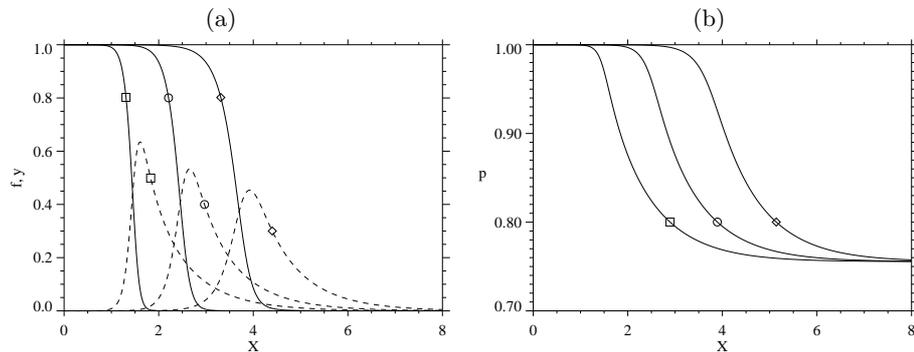


FIGURE 2. Steady detonation profiles showing (a) fuel, f , (solid lines) and radical, y , (dashed lines) and (b) pressure p . The symbols mark corresponding profiles for chain-branching cross-over temperatures $T_B = 0.80$ (\square), $T_B = 0.85$ (\circ), $T_B = 0.90$ (\diamond) and $T_B = 0.95$ (\triangle). Here $\theta_I = 20$, $\theta_B = 8$, $T_i = 3$, $q = 3$, $f = 1.2$ and $\gamma = 1.2$.

at the von-Neumann spike point of the steady wave are used for non-dimensionalisation. In practice, the initiation decomposition reaction is energetically inhibited, and typically proceeds at an exponentially small rate behind the detonation shock front. On the other hand, the branching reaction has a lower activation energy, involving reactions between energetic free radicals and atoms, and proceeds rapidly once a sufficient concentration of radicals has been established. Thus in typical, freely propagating, detonations the conditions

$$T_I > T_s > T_B, \quad \theta_I \gg \theta_B \gg 1, \quad (4.6)$$

where T_s is the shock temperature, would be expected to prevail. Finally the chemical energy q is defined as

$$q = Q(1 - f) - (Q + R)y, \quad (4.7)$$

where $Q > 0$ represents the total chemical energy available in the unreacted mixture and R represents the amount of endothermic energy absorbed by the initiation and chain-branching reactions in breaking down the reactant F into the energetic radical Y . In many cases, the endothermic energy R can be neglected, as in Short & Quirk (1997).

The most important bifurcation parameter in this model is that of the chain-branching cross-over temperature T_B . For other fixed parameters, it controls the ratio of the chain-branching induction region to that of the main heat release or chain-termination region. For values of T_B sufficiently below the von Neumann shock temperature, the rate of the chain-branching reaction will be sufficiently rapid that the chain-branching induction zone will be much shorter than the main reaction zone. On the other hand, if T_B is sufficiently close to the von Neumann shock temperature, the rate of chain-branching will be sufficiently small that the chain-branching induction zone will be much longer than the chain-termination region. Figure 2 shows the effect of varying T_B on the steady detonation wave structure. In Short & Dold (1996) and Short & Quirk (1997) the effect of varying T_B on the linear stability of detonations was ascertained. It was shown that planar detonations could in general be stabilized by decreasing T_B for both one- and two-dimensional disturbances. In particular, chain-branching detonations in which the temperature sensitive chain-branching induction is either longer or of the order of the length of the temperature-insensitive chain-termination region are unstable. Steady chain-branching detonations that are stable to one-dimensional perturbations occur in the limit that the chain-termination region is much longer than the chain-branching induction zone. The stability of detonations with the three-step reaction model has also

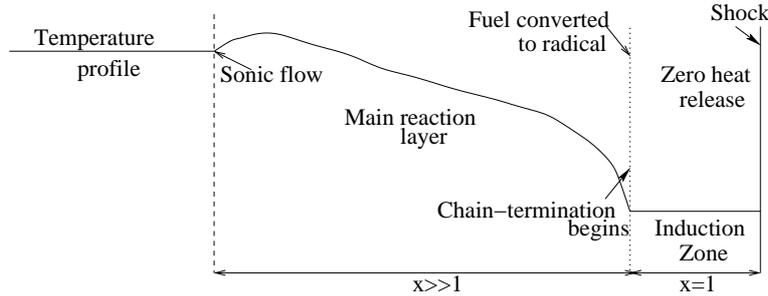


FIGURE 3. A schematic of the steady detonation structure for the two-step chain-branching reaction model.

been considered by Ng & Lee (2003). Recent work by Liang & Bauwens (2005) has extended this analysis to a four-step pressure-dependent chain-branching model.

A better handle on the ratio of the two length scales of chain-induction and chain-termination zones that lead to instability can be established by studying a two-step chain-branching like reaction model that is commonly used for detonation modeling (Sichel *et al.* 2002). Short (2001) considered a two-component model having a thermally neutral chain-branching induction zone and an exothermic main reaction layer (or chain-recombination layer) of finite extent, a schematic of which is shown in figure 3. The two layers are separated by a discontinuous transition layer, which in the three-step chain-branching model of Short & Quirk (1996) corresponds to a layer in which fuel is rapidly converted into chain radical. The rate equations may be written in the form

$$\frac{D\lambda_1}{Dt} = H(1 - \lambda_1)r_1, \quad \frac{D\lambda_2}{Dt} = (1 - H(1 - \lambda_1))r_2, \quad (4.8)$$

where λ_1 and λ_2 are the reaction progress variables in induction and main reaction layers respectively and $H()$ is the usual Heaviside function.

The dynamics in the induction zone are controlled by a reaction rate of Arrhenius form in which no heat is released due to reaction $q_1 = 0$, with

$$r_1 = k_1 \exp \left[\theta \left(\frac{1}{c_0^2} - \frac{1}{c^2} \right) \right], \quad (4.9)$$

where c_0 is the one-dimensional steady sound speed at the shock, $k_1 = O(1)$ is a rate constant and θ is the activation energy. At the shock $\lambda_1 = 0$, while the termination of the induction zone is signalled when $\lambda_1 = 1$, where fuel is instantaneously converted into chain-radical. The reaction rate in the exothermic chain-recombination layer ($q_2 = O(1) > 0$) is assumed to be independent of the local thermodynamic state, where

$$r_2 = k(1 - \lambda_2)^\nu, \quad (4.10)$$

Here k is the rate constant and ν is the reaction order. The size of k then determines the ratio of the length of the chain-recombination layer to the induction zone layer. For $k = O(1)$, the two layers are similar size, while for $k \ll 1$, the chain-recombination layer is much longer than the induction zone layer. Also, $\lambda_2 = 0$ marks the start of the main heat release layer, beyond which chain-recombination of the chain-radical occurs and heat is released, while $\lambda_2 = 1$ signals the rear equilibrium point of the detonation. An analysis of this rate model, shows that detonations which have $k < O(\theta^{-1})$ will be unstable to one-dimensional perturbations, and stable detonations require at least $k = O(1/\theta)$. This criteria is related to the work of Oppenheim, Meyer & Soloukhin on establishing the criteria between the ‘strong’ and ‘weak’ regimes of the ignition of gases

rapidly heated by the passage of a shock (Oppenheim 1985, Meyer & Oppenheim 1971). Radulescu (2003) has also demonstrated that a coherence relation between power pulses after a one-dimensional perturbation to a steady planar detonation wave leads to the result that stable detonations are generated when $k = O(1/\theta)$. Further refinements of this criteria have been proposed by Ng (2005).

4.3. Pathological detonation models

An interesting example of a series of reaction models that have important implications for detonation stability are those that lead to the so-called pathological detonation models (Fickett & Davis 1979). In this case, the speed of the self-sustaining detonation is characterised by a Rayleigh line being tangent to a Hugoniot curve corresponding to partial reaction, i.e. the sonic point occurs interior to the detonation. An example considered by Sharpe (1999) considers two consecutive, irreversible, reactions (fuel \rightarrow intermediate \rightarrow product). Consumption equations for fuel (f) and intermediate species (y) mass fractions are

$$Df/Dt = -r_1, \quad Dy/Dt = r_1 - r_2, \quad (4.11)$$

with rates

$$r_1 = \alpha_1 f \exp(-\theta_1/T), \quad r_2 = \alpha_2 y \exp(-\theta_2/T), \quad (4.12)$$

for activation energies θ_1 and θ_2 . The length scale (and hence α_1) is chosen such that $f = 1/2$ at $x = -1$ in the ZND wave. The heat release function q is

$$q = (q_1 + q_2)(1 - f) - q_2 y \quad (4.13)$$

where $q_1 > 0$ and q_2 are the heats of reaction in converting fuel to intermediate and intermediate to product respectively. When $q_2 < 0$, and provided that the rate the second is not sufficiently more rapid than the first, the sonic point appears in the detonation at a point of incomplete reaction. A normal mode stability analysis considered by Sharpe (1999) shows one qualitatively different behavior from the standard one-step Arrhenius reaction, namely that the detonation initially becomes more unstable as the overdrive is increased from one.

4.4. Summary

The study of the linear stability of steady detonation waves via a normal mode analysis is in a mature state. All issues regarding the appropriate closure conditions for both overdriven and Chapman-Jouguet waves of finite or infinite spatial extent have been resolved. In principle, we now possess the ability to calculate the neutral stability boundaries for steady detonations subject to multi-dimensional perturbations for any complex reaction mechanism and any equation(s) of state.

5. Comparison of linear predictions with DNS

The direct numerical simulation of detonations will be considered in a separate talk in this session given by Vadim Gamezo. We briefly remark here on the relation between the predictions of the linear stability analysis and direct numerical simulation of unstable detonations. This relation has been considered in a number of articles (e.g. Bourlioux, Majda & Roytburd 1991, Bourlioux & Majda 1992, He & Lee 1995, Short & Quirk 1997, Sharpe & Falle 1999, Sharpe & Falle 2000, Sharpe 2001, Short & Wang 2004, Henrick, Aslam & Powers 2004, Liang & Bauwens 2005). All the direct numerical simulations conducted thus far for one- and two-dimensional systems have verified the location of the neutral stability boundaries predicted by the linear analysis, and there appears to

be no evidence of sub-critical nonlinear bifurcation behavior. Also, for one-dimensional detonations that exhibit apparent nonlinear low-frequency (on the scale of the particle passage time through the steady reaction zone length) single-period limit cycle behavior, the frequency of the limit cycle is very close to that of the frequency associated with the lowest-frequency normal mode calculated from the linear analysis even for parameters far from those that determine the neutral stability point. This indicates that the basic mechanisms of the fully nonlinear one-dimensional evolution are captured by the linear analysis.

For two-dimensional simulations in a sufficiently narrow tube, the number of detonation cells observed in a channel with solid side walls corresponds well with the wavenumber of the most unstable mode obtained from the linear analysis that is compatible with the channel width (Bourlioux & Majda 1992, Sharpe 2001, Liang & Bauwens 2005). On the other hand, an analysis of two-dimensional calculations performed by Oran, Khokhlov & Gamezo, Bauwens, and Sharpe, Quirk & Short show that for wider tubes, the fully developed cells tend to have wavelengths that are several times larger than that predicted from the linear analysis and, moreover, the number of observed cells for a given parameter set tend to vary substantially with the form of the initial perturbation, at least for the length of time for which the calculations have been performed. Substantial work remains to be done in this area.

6. Asymptotic modeling of detonation stability

In order to gain insights into the physical mechanisms that drive the nonlinear mechanisms of detonation instability a number of asymptotic studies have been conducted, that either invoke a limiting decomposition of the underlying steady wave and/or make assumptions about the characteristic frequency and wavelength of the instability. Again, many of these studies tend to be somewhat technical in nature. Some of the main results are described below.

Yao & Stewart (1996) have derived a weakly nonlinear equation that describes the evolution of a near Chapman-Jouguet detonation wave in the limits (a) where the time-scale for the evolution is assumed to be slowly-evolving relative to the particle passage time through the steady wave, and (b) the front is weakly curved relative to the length scale of the steady reaction zone thickness. For a one-step reaction with a large activation energy, an explicit high-order evolution equation can be derived, which extends the earlier work of Buckmaster & Ludford (1996) by including higher-order correction terms in the limit of a low-frequency, long wavelength expansion. The result is a second-order evolution equation for the detonation shock velocity perturbation, which consists of a hierarchy of two-dimensional hyperbolic wave operators. Numerical solutions of the one-dimensional version of this equation demonstrate the presence of nonlinear limit cycle solutions. When the nonlinear terms that are responsible for the limit cycle generation are dropped, numerical solutions of the two-dimensional evolution equation generate cellular front patterns that are qualitatively similar to those observed in experiments (Stewart, Aslam & Yao 1997). One-dimensional evolution equations that govern the dynamics of CJ detonation waves for a two-step chain-branching-like reaction model with a spatially distributed main-heat release layer have also been derived in Short (2001) and Short & Sharpe (2003) assuming a low-frequency evolution.

An alternative approach for sufficiently overdriven waves has been described by Clavin & He (1996a). In this approach, the Mach number of the steady wave is assumed to be large, while the ratio of specific heats is assumed to be close to unity. When the heat release is taken to be of the order of the thermal enthalpy at the shock in these

limits, as occurs for sufficiently overdriven waves, the flow Mach number throughout the detonation will be small. The simplification that now arises in this limit is that to leading-order the flow behind the shock can be taken to be quasi-isobaric. In addition, by assuming that the reaction contains a large temperature state sensitivity at the shock, a nonlinear integral equation can be derived in the quasi-isobaric limit for small velocity departures from the steady detonation speed which is able to describe pulsating type of instabilities. Clavin & He (1996b) have included compressibility effects in this approach as a higher-order perturbation, where they are shown to have a stabilizing effect. Although in this high overdrive limit the instability mechanism is dominated by the fluctuations at the shock disturbing the heat release through their passage along entropy waves, and is not due to the amplification of acoustic disturbances by chemical energy release, Clavin (2004) suggests that the same mechanism will hold even for CJ detonations. Such an extrapolation seems hopeful at best, particularly for general state-dependent reaction rates, and the presence of thermoacoustic instabilities should not be eliminated based on the results of a limit which is non-uniform as the CJ wave speed is approached. Clearly more analysis is needed to identify the mechanisms of instability of CJ waves. Clavin, He & Williams (1997) have also studied the implications of sufficiently overdriven waves on the linear stability of detonations subject to multi-dimensional disturbances.

The limit of weak heat release, in which the heat release is smaller than the thermal enthalpy at the von Neumann shock state, has also been considered by several authors for overdriven waves (Short & Stewart 1999, He 2000, Short & Blythe 2002, Daou & Clavin 2003). This limit includes several cases, including finite $O(1)$ overdrives, and the limit of infinite overdrive for larger heat releases, together with a range of temperature sensitivities of the reaction rate at the shock. The simplification obtained in this limit is that the leading-order detonation structure is the uniform inert shock state. Short & Blythe (2002) have identified several distinguished limits between the small heat release, $(\gamma - 1) \ll 1$, and the inverse activation energy of the thermally-sensitive reaction at the shock that lead to instability. Clavin & Denet (2002) have also shown that numerical simulations of a weakly nonlinear integral-differential equation that may be derived in the limit of large overdrive induced weak heat release can qualitatively reproduce cellular front patterns. Finally, Clavin & Williams (2002) have also considered the limit of weak heat release for near CJ detonations.

In addition to these asymptotic approaches, more traditional one-dimensional and low-frequency two-dimensional (Ginzburg-Landau) weakly nonlinear evolution equations have been derived (Bourlioux, Majda & Roytburd 1991, Majda & Roytburd 1992).

7. Modeling of the stability of detonations in condensed phase explosives

In comparison to gas phase detonations, comparatively little is known about the reaction wave structure in detonating liquid and solid explosives, where the extreme high pressure environments (~ 30 - 50 GPa) and high wave speeds (~ 6 - 8 km/s) make experimental diagnostic data collection and imaging difficult. While there is some evidence that detonations in some forms of liquid explosives (such as nitromethane diluted with acetone) may exhibit a cellular structure, it has not yet been established whether this is due to an inherent reactive-hydrodynamic instability (as in gases) or due to the effects of explosive confinement (Fickett & Davis 1979). On the other hand, laser-based interferometry measurements of particle velocities in the reaction zones of detonations in pure and commercial grade liquid nitromethane (Sheffield *et al.* 2002), and in the solid explosives PBX9501 (Gustavsen, Sheffield & Alcon 2000) and PBX9502 (Seitz *et al.* 1989),

appear to indicate that the idealized ZND planar structure is stable. This conclusion is re-enforced by velocity against curvature measurements of detonations propagating in cylindrical sticks of nitromethane (Hill *et al.* 1999) and PBX9502 (Hill, Bdzil & Aslam 2000). However, the rapid time resolution ($\ll 1\text{ns}$) required to experimentally resolve the very fine structure of the detonation front in liquid and solid explosives is currently unavailable. Consequently it seems likely at this point in time that several advances in our understanding of detonation structure and stability in condensed phase systems will have to be made from mathematical and numerical modeling.

Much of the difficulty with the modeling of detonation propagation in condensed-phase explosives lies with our lack of knowledge of the equations of state for both partially reacted and product equations of state, and of the reaction mechanisms. Thus the modeling tends to be done within the context of empirical engineering scale models employing the continuum Euler equations of motion and, for example, assuming Mie-Gruneisen and JWL equations of state for reactants and products, with some mixture rule between reactants and products used to determine the partially reacted equations of state. In addition, many of the reaction models used in condensed phase detonation modeling assume pressure dependent rates. A popular example of this type of system is the Ignition and Growth (I&G) model (Tarver, Kury & Breithaupt 1997).

Short, Bdzil & Anguelova (2005) have recently examined the linear stability of condensed phase detonations with an Euler model for a one-step pressure-dependent reaction with a Tait (stiffened gas) equation of state, where

$$e = \frac{(p+a)\Lambda}{\Gamma_0} - \lambda q, \quad a = \delta - (\Gamma_0 + 1)p_0, \quad \delta = \frac{\tilde{c}_0^2}{\tilde{D}^2}, \quad p_0 = \frac{\tilde{\Lambda}_0 \tilde{p}_0}{\tilde{D}^2} \quad (7.1)$$

and

$$\frac{D\lambda}{Dt} = kp^n(1-\lambda)^\nu. \quad (7.2)$$

Here, $\delta^{-\frac{1}{2}}$ is the detonation Mach number with respect to the ambient sound speed, p_0 is the ratio of the ambient pressure \tilde{p}_0 to $\tilde{D}^2/\tilde{\Lambda}_0$, and Γ_0 is the Gruneisen gamma, which is assumed to be constant. Also, q ($= \tilde{q}/\tilde{D}^2$) is the non-dimensional heat of reaction. This system captures one of the (many) distinguishing features of condensed phase detonations in which the shock pressure p_s is substantially greater than the ambient pressure p_0 , but the detonation Mach numbers are typically in the range 2-4 due to the large ambient sound speed of the explosive. The value of a is in principle chosen to fit the experimentally determined sound speed in the material at a given pressure, where $c^2 = (1 + \Gamma_0)\Lambda(p + a/(1 + \Gamma_0))$. The values of δ and p_0 for the three condensed phase explosives NM and PBX9501/02 are listed in table 1 based on their material properties given in Marsh (1980). It can be seen that the contribution to a from p_0 is negligible, while the detonation Mach number $\delta^{-\frac{1}{2}}$ ranges from 3.78 in NM to 2.36 for PBX9502. When the sound speed $a = 0$, we recover the limit of the idealized condensed phase detonation model, which assumes an ideal gas equation of state with a large value of adiabatic gamma. Based on the values given in table 1, Short, Bdzil & Anguelova (2005) considered the parameter choices $p_0 = 0$, so $a = \delta$, with $0 \leq a \leq 0.2$. They also set $\nu = 1/2$ and $\Gamma_0 = 2$. For the Tait equation of state (7.1) and for a one-step irreversible reaction (7.2), the steady CJ detonation structure can be calculated exactly, and is given by

$$u = -\frac{(1+\Gamma)}{(2+\Gamma)}(1+\bar{p}_0) + \frac{(1-(1+\Gamma)\bar{p}_0)}{2+\Gamma}\beta^{1/2\mu}, \quad q = \frac{(1-(1+\Gamma)\bar{p}_0)^2}{2\Gamma(2+\Gamma)}, \quad (7.3)$$

where $\bar{p}_0 = p_0 + a/(1 + \Gamma)$.

	$\tilde{\rho}_0$ (g/cm ³)	\tilde{c}_0 (km/s)	\tilde{D}_{CJ} (km/s)	δ	p_0
NM	1.125	1.65	6.248	0.070	2.3×10^{-6}
PBX9501	1.844	2.96	8.792	0.113	7.1×10^{-7}
PBX9502	1.895	3.26	7.706	0.179	9.0×10^{-7}

TABLE 1. Characteristic properties of the explosives liquid nitromethane (NM), PBX9501 (95% HMX (cyclotetramethylenetetranitramine) and 5% binder by weight) and PBX9502 (95% TATB (triaminonitrobenzene), 5% Kel-F 800 by weight). Here $\tilde{\rho}_0$ and \tilde{c}_0 are the ambient material density and sound speed, while \tilde{D}_{CJ} is the characteristic planar Chapman-Jouguet detonation speed. All values are taken from Marsh (1980). The values of p_0 are calculated assuming $\tilde{p}_0 = 1$ atm.

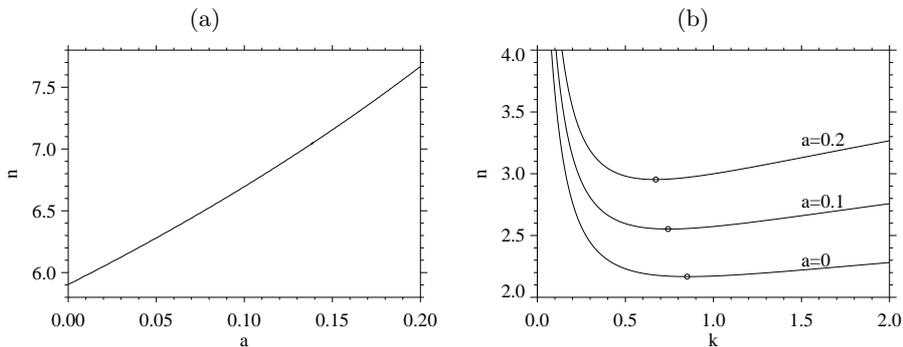


FIGURE 4. (a) One-dimensional neutral stability boundaries in the $n - a$ plane when $k_r = 1$. (b) Two-dimensional neutral stability boundaries in the $n - k$ plane ($k < 2$) for $a = 0$, $a = 0.1$ and $a = 0.2$ when $k_r = 1$. The circles mark the point of two-dimensional neutral stability in the (n, k) plane for each a .

Figure 1a shows the variation in the neutral stability boundary that governs one-dimensional ($k = 0$) stability as a varies. For the strong shock limit ($a = 0$), the neutral stability point occurs at $n = 5.904$. Increasing a has a three fold effect on the CJ wave structure: it reduces the effective heat release q , decreases the shock pressure, while lowering the overall length of the CJ wave. Correspondingly, as a increases, the value of the pressure exponent n at the point of one-dimensional neutral stability increases rapidly. Thus for fixed n , the CJ wave becomes more stable to one-dimensional disturbances as the detonation Mach number decreases. The neutral stability point based on table 1 for NM ($a = 0.07$) occurs at $n = 6.44$, for PBX9501 ($a = 0.113$) at $n = 6.81$ and for PBX9502 ($a = 0.179$) at $n = 7.44$. Figure 1b shows the neutral stability boundaries that govern stability to two-dimensional disturbances, and three values of a , namely $a = 0$, $a = 0.1$, and $a = 0.2$. Again, increasing a raises the value of the reaction rate pressure sensitivity exponent n required for instability. Instability when $a = 0$ occurs for $n > 2.168$, when $a = 0.1$ for $n > 2.552$ and when $a = 0.2$ for $n > 2.953$. Thus detonations in PBX9502 would be the most stable. Recent calculations of detonation stability in the I&G type systems indicates that the detonation waves are strongly stable, i.e. very large pressure exponent values are required for instability.

8. Future areas of analysis for detonation stability theory

There a number of outstanding issues in detonation wave structure and stability theory that remain to be resolved to further enhance of understanding of the dynamic evolution

of detonation waves, and which indicate some of the future directions the field of detonation stability theory should be taking. I should emphasize that these are the personal views of the author, and other researchers inevitably will have different opinions on the future directions for the field. The issues include

(i) *Linear stability of gaseous detonation waves with complex multi-step reaction mechanisms.* As noted in §2, we are now in a position to conduct such studies, e.g. for hydrogen/hydrocarbon/oxygen/diluent reactions, which thus far have only been conducted for model reduced-step chain-branching mechanisms. Indeed, there have a number of recent papers that have attempted to conduct direct numerical simulations of detonation wave propagation for complex multi-step reaction mechanisms (e.g. Radulescu *et al.* 2002, Yungster & Radhakrishnan 2004, 2005, Ng 2005), although issues remain as to our current ability to adequately resolve the reaction zone structure for such mechanisms. In the context of such studies, it will be of substantial interest to analyze what particular mechanisms and species are essential to the development of detonation instability. Such calculations could also be of use for comparisons with recently developed PLIF experimental methods for tracing individual species profiles behind the cellular front of a detonation (e.g. Austin, Pintgen & Shepherd 2005).

(ii) *Linear and weakly nonlinear stability analysis of detonation evolution under engineering scale models of condensed-phase explosives.* As noted in §7, comparatively little is known about fine-scale detonation wave structure in liquid and solid explosives. In order to gain some insights into how the properties of detonations in condensed-phase systems differ from those in gas-phase systems, it seems that a reasonable starting point should be an analysis of the linear and weakly nonlinear stability behavior of condensed-phase detonations using the empirical continuum engineering-scale models most commonly used for studying condensed-phase systems. Some initial work in this area has been conducted by Short *et al.* 2005, Short, Bdzil & Anguelova (2005) & Kasimov (2004), but the area is ripe for exploration via a number of different strategies and models.

(iii) *Asymptotic analysis of the evolution of weakly nonlinear cellular CJ waves:* Detonations of the most practical importance in either gaseous or condensed phase explosives are those that run with speeds close to CJ. Like most things in nature, the most important cases are often the most difficult to analyse, and this is certainly the case here. In addition to important limiting cases, such as that of weak heat release (Clavin & Williams 2002) or low-frequency evolutions (Yao & Stewart 1996, Short 2001), there is a substantial need for asymptotic approaches to the weakly nonlinear evolution of CJ detonations that impose more practical limits. However, several severe technical issues must then be dealt with, such as accounting for the multi time-scale nature of various thermo-acoustic processes on-going in the evolution, as well as the likely generation of forward-propagating waves from the sonic equilibrium zone generated by resonant wave interactions (Bdzil & Klein 1992). In any case, any progress in this area will be very valuable.

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