Asymptotic Study of Pulsating Evolution of Overdriven and CJ Detonation with a Chain-Branching Kinetics Model

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1   Introduction

The pulsating dynamics of gaseous detonations with a model two-step chain-branching kinetic mechanism are studied both numerically and asymptotically. The model studied here was also used in [4], [3] and [2] and mimics the attributes of some chain-branching reaction mechanisms. Specifically, the model comprises a chain-initiation/branching zone with an Arrhenius temperature-sensitive rate behind the detonation shock where fuel is converted into chain-radical with no heat release. This is followed by a chain-termination zone having a temperature insensitive rate where the exothermic heat of reaction is released. The lengths of these two zones depend on the relative rates of each stage. It was determined in [4] and [3] via asymptotic and numerical analysis that the ratio of the length of the chain-branching zone to that of the chain-initation zone relative to the size of the von Neumann state scaled activation energy in the chain initiation/branching zone has a primary influence on the stability of one-dimensional pulsating instability behavior for this model. In [2], the notion of a specific stability parameter related to this ratio was proposed that determines the boundary between stable and unstable waves.

In [4], a slow time-varying asymptotic study was conducted of pulsating instability of Chapman-Jouguet (CJ) detonations with the above two-step rate model, assuming a large activation energy for the chain-initiation zone and a chain-termination zone longer than the chain-initiation zone. Deviations $D_n^{(1)}(\tau)$ of the detonation velocity from Chapman-Jouguet were of the order of the non-dimensional activation energy. Solutions were sought for a pulsation timescale of the order of the non-dimensional activation energy times the particle transit time through the induction zone. On this time-scale, the evolution of the chain-initation zone is quasi-steady. A time-dependent non-linear evolution equation for $D_n^{(1)}(\tau)$ was then constructed via a perturbation procedure for cases where the ratio of the length of the chain-termination zone to chain-initiation zone was less than the non-dimensional activation energy. To leading order, the steady CJ detonation was found to be unstable; higher-order corrections lead to the construction of a stability limit between stable and unsteady pulsating solutions. One conclusion from this study is that for a stability limit to occur at leading order, the period of pulsation of the detonation must occur on the time scale of particle passage through the longer chain-termination zone, while the length of the chain-termination zone must be of order of the non-dimensional activation energy longer than the

chain-initiation zone. The relevance of these suggested scalings was verified via numerical solutions of the full Euler system in [3], and formed the basis of the stability parameter criteria suggested in [2]. In the following, we formulate an asymptotic study based on these new suggested scales, studying the implications for describing pulsating behavior in gaseous chain-branching detonations. Specifically, we find that the chain-induction zone structure is the same as that studied in [4]. However, the study of unsteady evolution in the chain-termination region is now governed by a set of asymptotically derived nonlinear PDEs. Equations for the linear stability behavior of this set of PDE's are obtained, while the nonlinear PDEs are solved numerically using a shock-attached, shock-fitting method developed by Henrick *et al.* [1]. The results thus far show that the stability threshold calculated using the new ratio of the chain-termination zone length to that of the chain-initiation zone yields a marked improvement over [4]. Additionally, solutions will be compared with predictions obtained from the solution of the full Euler system. Finally, the evolution equation previously derived in [4] has been generalized to consider both arbitrary reaction orders and any degree of overdrive.

2 Detonation model and equations

The reactive Euler equations are used to model the evolution of the perturbations to the fluid density, velocity, pressure, reaction progress, and to the shock velocity. These are,

$$\frac{D\rho}{Dt} + \rho\nabla \cdot \mathbf{u} = 0,\ \frac{D\mathbf{u}}{Dt} + \frac{1}{\rho}\nabla p = 0,\ \frac{De}{Dt} + p\frac{D}{Dt}\left(\frac{1}{\rho}\right) = 0$$

$$\frac{D\lambda_i}{Dt} = r_i \quad \text{where} \quad \begin{cases} r_1 = k_1 H(1-\lambda_1)\exp\left(E\left(\frac{1}{RT_0} - \frac{1}{RT}\right)\right) \\ r_2 = k_2(1 - H(1-\lambda_1))(1-\lambda_2)^\nu \end{cases} \tag{1}$$

where the two-stage reaction is parametrized by the reaction progress variables λ_1 and $\lambda_2 \in [0,1]$. Here, T_0 denotes the post-shock temperature for the steady wave, and R is the gas constant. This particular form for the reaction rates ensures that the induction zone is characterized by a single reaction. Furthermore, the model assumes an ideal gas equation of state and specific internal energy of the form

$$\frac{p}{\rho} = RT,\ e = \frac{1}{\gamma - 1}\frac{p}{\rho} - \sum_i Q_i\lambda_i \tag{2}$$

which leads to $c^2 = \gamma p/\rho$, wehere γ is the specific heats ratio.

In a frame moving with the steady CJ detonation ($n = x - Dt$, $u \to u_n + D$), these equations can be written as

$$\frac{\partial\rho}{\partial t} + \frac{\partial}{\partial n}\left(\rho u_n\right) = 0,\ \frac{\partial}{\partial t}(\rho u) + \frac{\partial}{\partial n}\left(\rho u_n^2 + p\right) = 0,\ \frac{\partial\lambda}{\partial t} + u\frac{\partial\lambda_i}{\partial n} = r_i$$

$$\frac{\partial u}{\partial t} + \frac{1}{u}\left(\frac{1}{(\gamma-1)\rho}\frac{\partial p}{\partial t} - \frac{\gamma p}{(\gamma-1)\rho^2}\frac{\partial\rho}{\partial t} - \sum_i Q_i\frac{\partial\lambda_i}{\partial t}\right) + \frac{\partial}{\partial n}\left(\frac{\gamma}{\gamma-1}\frac{p}{\rho} + \frac{u_n^2}{2} - \sum_i Q_i\lambda_i\right) = 0 \tag{3}$$

The steady state structure for the two stages of reaction is obtained through the conservation relations,

$$\frac{\partial}{\partial n}\left(\rho u_n\right) = 0,\ \frac{\partial}{\partial n}\left(\rho u_n^2 + p\right) = 0,\ \frac{\partial}{\partial n}\left(\frac{\gamma}{\gamma-1}\frac{p}{\rho} + \frac{u_n^2}{2} - \sum_i \beta_i\lambda_i\right) = 0,\ \frac{\partial\lambda_i}{\partial n} = \frac{r_i}{u_n} \tag{4}$$

Although in Ref. [4], a non-dimensional reference system is based on steady post-shock quantities, here a system of reference is built upon the CJ propagation velocity, D_{CJ}, and the pre-shocked density, ρ_u. Additionally, the relevant length scale is the length of the induction zone, l_I, and the time scale is l_I/D_{CJ}. The scalings are summarized as,

$$\rho = \frac{\tilde{\rho}}{\tilde{\rho}_u}, \; U_n = \frac{\tilde{U}_n}{\tilde{D}_{CJ}}, \; p = \frac{\tilde{p}}{\tilde{\rho}_u \tilde{D}_{CJ}^2}, \; T = \frac{\tilde{T}}{\tilde{D}_{CJ}^2/(\gamma \tilde{R})}, \; \beta_i = \frac{\tilde{Q}_i}{D_{CJ}^2},$$

$$\epsilon = \frac{1/\tilde{E}}{\gamma \tilde{R}/\tilde{D}_{CJ}^2}, \; x = \frac{\tilde{x}}{\tilde{l}_I}, \; t = \frac{\tilde{t}}{t_I} = \frac{\tilde{t}}{\tilde{l}_I/\tilde{D}_{CJ}} \tag{5}$$

The unannoted symbols above from this point forward refer to dimensionless quantities for ease of notation. Additionally, the tilde appearing above a quantity will henceforth denote a dimensional quantity in order to differentiate between dimensional and non-dimensional quantities.

3 Asymptotic formulation

We examine the dynamics of pulsating detonation withing the framework of the two-step model, assuming a small non-dimensional activation energy in the chain-initiation zone. A perturbation in the shock velocity occurs in the form

$$D_n = D_n^{(0)} + \epsilon D_n^{(1)}(\tau) \tag{6}$$

where ϵ is the inverse activation energy and $\epsilon \ll 1$. $D_n^{(0)}$ is the constant steady detonation velocity, with unsteady perturbations occuring on the slow time scale $\tau = \epsilon t$. The reason for the choice of the slow time scale of $O(1/\epsilon)$ is explained below. The new shock position, $h(t,\tau)$, is $n = x - h(t,\tau)$, where $h(t,\tau) = D_n^{(0)}t + H(\tau)$. In the perturbed shock frame, the Euler equations are

$$\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial n}\big((u_n - \epsilon D_n^{(1)})\rho\big) = 0, \; \frac{\partial}{\partial t}(\rho u_n) + \frac{\partial}{\partial n}\big(\rho(u - \epsilon D_n^{(1)})u_n + p\big) = 0$$

$$\frac{1}{u_n - \epsilon D_n^{(1)}}\left(\frac{\partial e}{\partial t} - \frac{p}{\rho^2}\frac{\partial \rho}{\partial t}\right) + \frac{\partial u}{\partial t} + \frac{\partial}{\partial n}\left(\frac{u_n^2}{2} + e + \frac{p}{\rho}\right) = 0, \; \frac{1}{u_n - \epsilon D_n^{(1)}}\frac{\partial \lambda}{\partial t} + \frac{\partial \lambda}{\partial n} = \frac{r_i}{u_n - \epsilon D_n^{(1)}} \tag{7}$$

In the chain-initiation zone, each quantity is expanded in powers of ϵ, i.e.

$$\rho = \rho^{(0)} + \epsilon \rho^{(1)} + O(\epsilon^2), \; u_n = u_n^{(0)} + \epsilon u_n^{(1)} + O(\epsilon^2), \; p = p^{(0)} + \epsilon p^{(1)} + O(\epsilon^2),$$

$$\lambda_i = \lambda_i^{(0)} + \epsilon \lambda_i^{(1)} + O(\epsilon^2) \tag{8}$$

where the leading order quantities are constant and corresponding to the post-shock von Neumann state in either the CJ or overdriven steady wave. Expanding the Rankine-Hugoniot conditions at the shock using (8) leads to the boundary condition that at $n = 0$,

$$u_{n0}^{(1)} = k_{u_n} D_n^{(1)}, \quad p_0^{(1)} = k_p D_n^{(1)}, \quad \rho_0^{(1)} = k_\rho D_n^{(1)}. \tag{9}$$

As there is no heat release in the chain-induction zone, these perturbations are transmitted through the chain-initiation zone without change in form, and define the $O(\epsilon)$ amplitude conditions at the interface between the chain-initiation/branching and chain-termination zones. The motion of the aforementioned

interface as a result of time-dependent perturbations at the shock is determined by expanding $T = c^2 = \gamma p/\rho$, giving

$$\frac{\partial \lambda_1^{(0)}}{\partial n} = \frac{r_1^{(0)}}{u_n^{(0)}} \text{ with } r_1 = k_1 \exp\left(\frac{1}{\epsilon}\left(\frac{1}{T_0} - \frac{1}{T}\right)\right) \Rightarrow \frac{\partial \lambda_1^{(0)}}{\partial n} = -\exp\left(\frac{\rho^{(0)} p^{(1)}}{\gamma (p^{(0)})^2} - \frac{\rho^{(1)}}{\gamma p^{(0)}}\right) \quad (10)$$

Consequently, the $O(\epsilon)$ perturbations in the chain-initiation zone have an $O(1)$ effect on the reaction progress variable spatial derivative, $\partial \lambda_1^{(0)}/\partial n$, which in turn has an order $O(1)$ affect on the location of the interface between the chain-termination and induction zones. Explicitly, one obtains that the induction zone length (relative to the shock) is $F(\tau) = -\exp(bD_n^{(1)})$. When viewed on the slow time scale τ, the time-dependent changes in the location of the chain-initiation/chain-termination interface lead to the introduction of $O(\epsilon)$ perturbations into the chain-branching zone. Since the amplitude of these perturbations must be balanced by the $O(\epsilon)$ perturbations transmitted through the chain-initation zone from the detonation shock, it underlies the choice of the time variable $\tau = \epsilon t$.

The analysis of the corresponding evolution of the chain-termination zone proceeds by changing the frame of reference to the chain-initiation/chain-termination interface, i.e.

$$m = (\epsilon/\omega)(n - F(\tau)). \quad (11)$$

Seeking solutions (8), a set of PDE's is derived at $O(\epsilon)$ in which the acoustic terms are linearized but have spatially varying coefficients, while nonlinearity enters through the motion of the interface ($m = 0$). One can reform these PDEs into a matrix form (the form of the matrices are omitted for brevity) by defining $\mathbf{v}^{(1)} = (\rho^{(1)}, u^{(1)}, p^{(1)}, \lambda^{(1)})^T$, i.e.

$$\omega A \frac{\partial \mathbf{v}^{(1)}}{\partial t} + \frac{\partial}{\partial m}(B\mathbf{v}^{(1)}) + C\mathbf{v}^{(1)} - (D_n^{(1)} + F_\tau)\mathbf{g} = 0, \quad (12)$$

that are subject to

$$\mathbf{v}^{(1)}(m = 0, \tau) = (k_\rho, k_{u_n}, k_p, 0)^T \times D_n^{(1)}. \quad (13)$$

For overdriven waves, the system is closed through an ansatz of an acoustic radiation condition downstream of the detonation. CJ detonation waves require special attention due the introduction of a further slow time variable in a transonic zone near the end of the detonation wave. The parameter ω is essential to the analysis here and in [4], [3] and [2]. As mentioned earlier, a well-defined stability problem requires that the chain-termination zone be longer than the chain-initiation zone. This introduces a rescaling of the rate constant in the chain-termination zone to a new modified rate constant defined by $\tilde{k} = \omega k_2/\epsilon$. If $\epsilon \ll \omega \ll 1$, the pulsation frequency of the instability is long on the scale of particle transit time through the chain-termination zone. With this assumption, [4] and [3] conducted a perturbation analysis of the behavior of the chain-termination zone via the expansion $\rho^{(1)} \rightarrow \rho^{(1)} + \omega \rho^{(2)}$. However, the leading-order problem only possesses unstable solutions. In the following, we have extended the analysis in [4] and [3] to arbitrary reaction orders in the chain-termination zone as well as to overdriven waves. Generally, though, in order to recover a stability limit in the leading order problem, the ratio of the length of the chain-termination zone to that of the chain-initiation zone must be $O(1/\epsilon)$, i.e. by taking $\omega = O(1)$. The main purpose of the present work is to solve and analyze the PDE system (12), describing pulsating instabilities of the two-step chain-branching detonation.

4   Generalized slow varying evolution equation

As noted above, the evolution equation derived in [4] can be generalized to any order ν and overdrive. The evolution equation is obtained via a regularity condition that is imposed at the end of the chain-termination zone. In the CJ case, the perturbations have an explicitly non-regular component that is
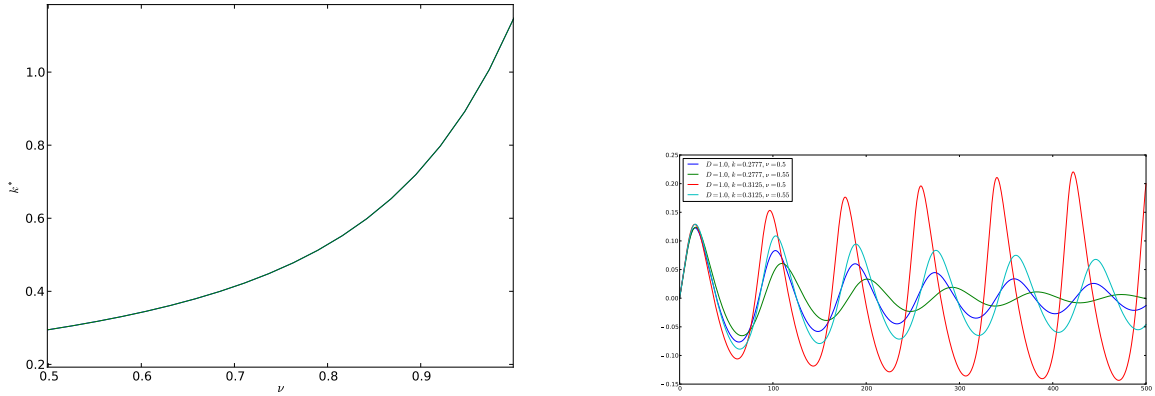
Figure 1. (Left) The threshold value of k^* defined the boundary between stable and unstable solutions as a function of the reaction order ν for CJ detonation. (Right) The effect of increasing the reaction order to $\nu = 1$ in the solution obtained for $D_n(\tau)$ for various values of k.

eliminated by imposing a certain relation between D_n and its time derivative. One can generalize this by imposing a radiation condition for overdriven waves. It can be easily shown that this is equivalent in the limit as $D \to 1$ to the regularization condition imposed by Short [4]. Typical results for the overdriven case and for varying reaction orders are shown in figs. 1 and 2.

5   Numerical solution of the perturbation PDE system

The PDE system (12) can be solved subject to the imposition of an initial perturbation to obtain the subsequent behavior of $D_n^{(1)}$ as a function of τ. We have chosen a solution strategy based on the shock-fit, shock-attached approach derived by Henrick *et al.* [1]. The behavior of $D_n^{(1)}(\tau)$ is obtaining through the so called "shock change" equation at $m = 0$. The particular scheme used here is a centered 4th order spatial scheme in the interior of the computational domain and one-sided differences near the boundaries at $m = 0$ and at the end of the reaction zone. A 5th order temporal Runge-Kutta routine is used to integrate in time. Figure 3 shows a preliminary calculation of $D_n^{(1)}(\tau)$ calculated using this method for a stable case with $Q = 4.0$, $\gamma = 1.4$, $\nu = 0.5$ and $D = 1$. The stability threshold obtained directly via the PDE system is found to be $\tilde{k} = 0.415$. In [3], the stability threshold was found to be $\tilde{k} = 0.43$ obtained via a full numerical solution of the Euler system, while [3] also predicted $\tilde{k} = 0.29$ for the slowly varying evolution equation analysis (where $\omega \ll 1$).

A formal linear stability analysis of the system (12) has also been conducted via a normal mode analysis. The full paper will provide results from this analysis to be compared with the numerical solution of (12). The work sheds significant light on the ratio of length and time scales and their relationship to a basic model of gas-phase chain-branching reaction kinetics that underlie pulsating detonation behavior.

References

[1] A.K. Henrick, T.D. Aslam, and J.M. Powers. Simulations of pulsating one-dimensional detonations with true fifth order accuracy. *Journal of Computational Physics*, 213:311–329, 2006.
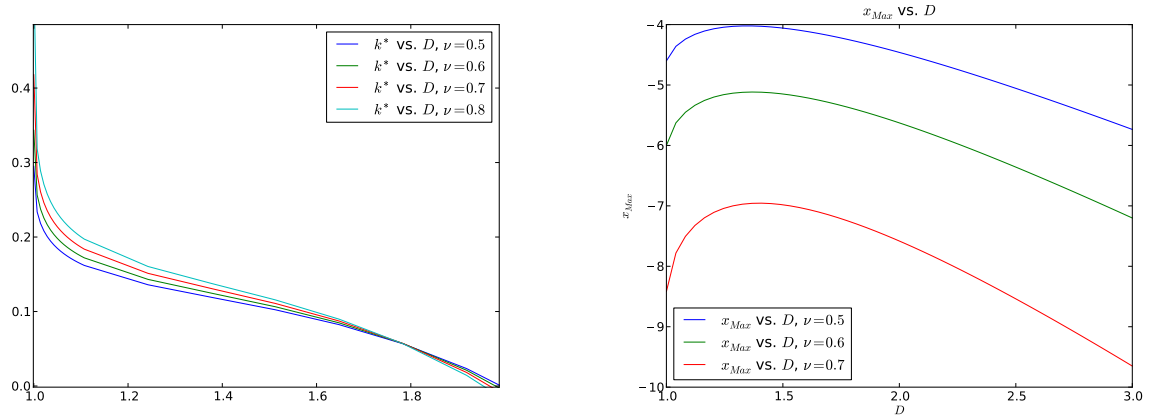
Figure 2. (Left) The effect of increasing overdrive D for various values of ν on the stability threshold in $\tilde{k}$. (Right) The reaction zone length plotted versus overdrive D. There is a range of values for which the CJ detonation length is longer than the overdriven case. This helps explain the increased instability range in $\tilde{k}$.
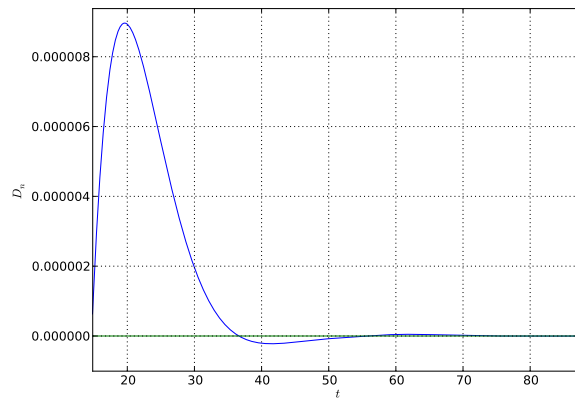

Figure 3. The evolution of the detonation velocity perturbation for $\tilde{k} = 0.41$ for a decaying perturbation.

[2] H.D. Ng, M.I. Radulescu, A.J. Higgins, N. Nikiforakis, and J.H.S. Lee. Numerical investigation of the instability for one-dimensional Chapman-Jouguet detonations with chain-branching kinetics. *Combust. Theory and Modell.*, 9:385–401, 2005.

[3] M. Short and G.J. Sharpe. Pulsating instability of detonations with a two-step chain-branching reaction model: theory and numerics. *Combution Theory and Modeling*, 7:401–416, 2003.

[4] Mark Short. A nonlinear evolution equation for pulsating Chapman-Jouguet detonations with chain-branching kinetics. *Journal of Fluid Mechanics*, 430:381–400, 2001.