

Linear stability of detonations with reversible chemical reactions

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1 Introduction

Experiments and numerical simulations of detonation propagation problems are characterized by unsteady motion due to the intrinsic instability of the reaction zone structure. Investigations of the detonation linear stability problem have almost exclusively been concerned with model systems using the perfect gas equation of state and reaction mechanisms consisting of a small number (usually one) of irreversible reactions [1]. Starting from the one-step model used in the pioneering studies of Erpenbeck and the reformulation of the numerical approach by Stewart and coworkers [2, 3, 4], researchers have been making steady progress by considering more complex chemical reaction models and equations of state [5, 6].

The scope of our present study is the linear stage of the instability of an initially steady, one-dimensional base flow based on the ZND model of detonation structure. Our goal is to develop numerical tools that will enable the computation of stability characteristics for multi-step reaction mechanisms with reversible reactions, realistic rate constant representation, and mixture thermodynamics with variable specific heat. To this end, we have generalized the formulation of the linear stability equations following the method of Lee and Stewart [2] and Short [3, 4].

We have verified that our method will reproduce the previous one- and two-dimensional results of Lee and Stewart and Short and Stewart and have proceeded to examine the effect of reversibility on the one-step reaction. We have constructed a family of CJ solutions with a fixed CJ temperature but varying extents of reversibility in the reaction rate. The extent of reversibility is controlled by the entropy of the product state relative to the reactant. Using this family of CJ solutions, we have computed the unstable modes as a function of the extent of reversibility and degree of overdrive.

2 Formulation and Implementation

The system of interest is governed by the reactive Euler equations. Following the normal modes approach outlined in Lee and Stewart [2] and Short [3], the generalized dimensional governing equations are the ZND equations and

$$\mathbf{A}^o \mathbf{z}_{,x}^1 + (\omega \mathbf{I} + \mathbf{C}^o + ik \mathbf{B}^o) \mathbf{z}^1 + (\omega \mathbf{I} + ik \mathbf{B}^o) \mathbf{b}^o \psi^1 = 0 \quad (1)$$

where

$$\mathbf{z} = [v, u, v, P, Y_1, Y_2, \dots, Y_N]^T \quad Z(P, v, \mathbf{Y}) = -\frac{G}{v} \sum_{i=1}^N e_{Y_i} \dot{\Omega}_i \quad \mathbf{b} = \mathbf{z}_{,x} \quad (2)$$

$$\mathbf{A} = \begin{pmatrix} u & -v & 0 & 0 & 0 & 0 & \dots \\ 0 & u & 0 & v & 0 & 0 & \dots \\ 0 & 0 & u & 0 & 0 & 0 & \dots \\ 0 & \rho a_f^2 & 0 & u & 0 & 0 & \dots \\ 0 & 0 & 0 & 0 & u & 0 & \dots \\ 0 & 0 & 0 & 0 & 0 & u & \dots \\ \dots & & & & & & \end{pmatrix} \quad \mathbf{B} = \begin{pmatrix} v & 0 & -v & 0 & 0 & 0 & \dots \\ 0 & v & 0 & 0 & 0 & 0 & \dots \\ 0 & 0 & v & v & 0 & 0 & \dots \\ 0 & 0 & \rho a_f^2 & v & 0 & 0 & \dots \\ 0 & 0 & 0 & 0 & v & 0 & \dots \\ 0 & 0 & 0 & 0 & 0 & v & \dots \\ \dots & & & & & & \end{pmatrix} \quad (3)$$

$$\mathbf{C} = \begin{pmatrix} -u_{,x} & v_{,x} & 0 & 0 & 0 & 0 & \dots \\ P_{,x} & u_{,x} & 0 & 0 & 0 & 0 & \dots \\ 0 & v_{,x} & 0 & 0 & 0 & 0 & \dots \\ (\rho a_f^2)_{,v} u_{,x} - Z_{,v} & P_{,x} & 0 & (\rho a_f^2)_{,P} u_{,x} - Z_{,P} & (\rho a_f^2)_{,Y_1} u_{,x} - Z_{,Y_1} & (\rho a_f^2)_{,Y_2} u_{,x} - Z_{,Y_2} & \dots \\ -\dot{\Omega}_{1,v} & Y_{1,x} & 0 & -\dot{\Omega}_{1,P} & -\dot{\Omega}_{1,Y_1} & -\dot{\Omega}_{1,Y_2} & \dots \\ -\dot{\Omega}_{2,v} & Y_{2,x} & 0 & -\dot{\Omega}_{2,P} & -\dot{\Omega}_{2,Y_1} & -\dot{\Omega}_{2,Y_2} & \dots \\ \dots & & & & & & \end{pmatrix} \quad (4)$$

The superscripts in Equation 1 designate the base state (o) and the perturbed state (1), ψ^1 is the constant magnitude of the initial perturbation, ω is the complex eigenvalue, and k is the wave number. The other symbols represent: v specific volume, (u, v) normal and tangential velocity, P pressure, $\mathbf{Y}$ species, $\dot{\Omega}_i$ net production rate of species i , ρ density, a_f frozen soundspeed, e internal energy, and G Grüneisen coefficient. In addition to these equations, we use the ideal gas equation of state.

We have implemented the solution in the physical space domain as a shooting problem that requires the solution of the set of coupled ordinary differential equations for the base flow and perturbed quantities. Our implementation uses Cantera [7] to evaluate all thermodynamic quantities and derivatives of quantities dependent on the kinetics model. By using Cantera, the user is free to select the mechanism, thermodynamic, and reaction rate models specific to his problem. This includes the ability to use the sizable hydrocarbon fuel reaction mechanisms that have been developed by the combustion community in the last two decades as well as reduced or notional mechanisms with pseudo-species.

In previous implementations, the one step irreversible model [2] has been hard-coded and the user provides several parameters. Most researchers have validated their implementations using Erpenbeck's case: non-dimensional activation energy $\tilde{E} = 50$, non-dimensional heat release $\tilde{Q} = 50$, ratio of specific heats $\gamma = 1.2$, and overdrive $f = (U/U_{CJ})^2 = 1.2$. Using Cantera, we are able to create an input file that mimics this case. In addition, we have the flexibility to specify a reversible reaction and vary the amount of reversibility, i.e. the amount of product achieved at equilibrium, by adjusting the entropy difference between the reactant and product species. We have chosen to create a family of systems each with one global reversible reaction where the Chapman-Jouguet temperature T_{CJ} remains constant. To achieve this we specify the desired entropy difference ($\Delta S/\mathcal{R}$) and desired T_{CJ} and solve for the required heat release ($\Delta H/(\mathcal{R}T_0)$). Table 1 describes the family of systems that we have created. We see that the $\Delta S/\mathcal{R} = 0$ case is comparable to the irreversible case.

The computational domain is the reaction zone in the shock-fixed frame such that the left boundary conditions are the perturbed shock jump conditions. We have derived these for a general equation of state and implemented them for an ideal gas equation of state. At the right boundary, the system must satisfy a radiation condition requiring that all waves travel out of the domain. In the case of a single reversible reaction between perfect gases with equal molecular weight, the characteristic wave speeds are

$\Delta S/\mathcal{R}$	$\Delta H/(\mathcal{R}T_0)$	U_{CJ}	λ_{eq}
0	50.70	1701.15	0.986
-1	51.74	1700.30	0.965
-2	53.93	1699.01	0.924
-3	57.78	1697.56	0.860
-4	63.35	1696.42	0.783
-5	70.34	1695.66	0.703
-6	78.38	1695.20	0.630
-7	87.17	1694.90	0.566
-8	96.50	1694.72	0.511

Table 1: Reversibility parameters for $T_{CJ} = 3599.29$ K

complex and can be determined by the following polynomial.

$$-\omega\tau_o^* \left(\frac{a_e}{a_f}\right)^2 [c - u_o] [(c - u_o)^2 - a_f^2] + c [(c - u_o)^2 - a_e^2] = 0 \quad (5)$$

where a_f and a_e are the frozen and equilibrium soundspeeds, c is the characteristic wave speed, and τ^* is a function of the *near equilibrium relaxation constant*, $\tau = \left(\frac{\partial \dot{\Omega}}{\partial \lambda}\right)^{-1}$. The roots of this equation are depicted in Figure 1. The boxes indicate the limiting behavior of the irreversible reaction when $a_f = a_e$. We have noted that only one root consistently has a negative real part and radiates back into the system.

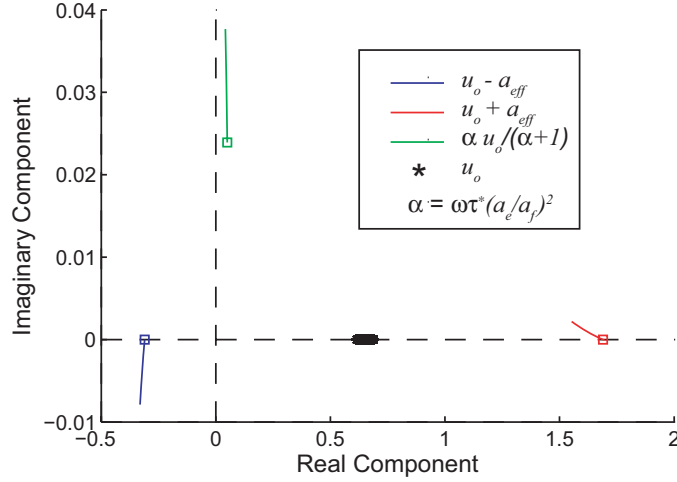


Figure 1: Roots of Eq. 5 for the first mode [2].

We have shown that determining the eigenvalues of the following representation of our problem is equivalent to determining the characteristic speeds.

$$\frac{1}{c} \mathbf{z}^1 = \mathbf{A}^{-1} \left[\mathbf{D} - \frac{ik}{\omega} \mathbf{B} - \frac{1}{\omega} \mathbf{C} \right]^o \mathbf{z}^1 \quad (6)$$

where

$$\mathbf{D} = \begin{pmatrix} 1 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 \\ -a_f^2 & 0 & 0 & 1 & -a_f^2 \rho_o \sigma \\ 0 & 0 & 0 & 0 & 1 \end{pmatrix} \quad (7)$$

We then dot the numerically determined eigenvector corresponding to this eigenvalue with the solution vector far from the reaction zone to determine a numerical radiation condition. This alternate methodology applied to the one dimensional irreversible system results in a radiation condition identical to that presented in [2]. We can also neglect the chemistry (i.e. $\mathbf{C} = 0$) and recover the condition presented in [3] for the two dimensional irreversible system. The shooting method ensures that the radiation boundary condition is satisfied within a user-specified tolerance. In a large multi-step mechanism with reversible reactions, we believe that this method will be applicable.

Another important consideration concerning the Cantera input files is the length scale of the problem. We implement the problem using the CVODE solver [11] and must non-dimensionalize pressure and velocity quantities to appease the solver. For this reason, we adopt the half reaction length as the length scale [2, 3] and the post-shock pressure as the pressure scale. This length scale is imposed by adjusting the pre-exponential component of the Arrhenius reaction rate such that $\lambda = \lambda_{eq}/2$ occurs at $t = 1$. Due to this requirement, the pre-exponential value varies with overdrive f and degree of reversibility $\Delta S/\mathcal{R}$.

3 Work in Progress

Now that we have created the family of input families described above, we plan to investigate the effects of reversibility on stability as a function of overdrive. We will look at differences between using a frozen radiation condition ($\mathbf{C} = 0$) and a reactive radiation condition ($\mathbf{C} \neq 0$). Eventually, we plan to study this problem in two dimensions ($k \neq 0$) and for more complex chemical mechanism models.

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