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A Kinetic Approach to Propagation and Stability of Detonation Waves

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Abstract. The problem of the steady propagation and linear stability of a detonation wave is formulated in the kinetic frame for a quaternary gas mixture in which a reversible bimolecular reaction takes place. The reactive Euler equations and related Rankine-Hugoniot conditions are deduced from the mesoscopic description of the process. The steady propagation problem is solved for a Zeldovich, von Neuman and Doering (ZND) wave, providing the detonation profiles and the wave thickness for different overdrive degrees. The one-dimensional stability of such detonation wave is then studied in terms of an initial value problem coupled with an acoustic radiation condition at the equilibrium final state. The stability equations and their initial data are deduced from the linearized reactive Euler equations and related Rankine-Hugoniot conditions through a normal mode analysis referred to the complex disturbances of the steady state variables. Some numerical simulations for an elementary reaction of the hydrogen-oxygen chain are proposed in order to describe the time and space evolution of the instabilities induced by the shock front perturbation.

Keywords: Boltzmann equation, Chemical reactions, Steady detonation solution, Linear stability
PACS: 47.15.Fe 47.45.Ab 47.70.Fw 82.33.Vx

INTRODUCTION

In some recent papers [1, 2] the problem of the steady propagation of a detonation wave has been considered starting from a physical system represented by a reactive Boltzmann equation for reversible bimolecular chemical reactions. Such papers indicate a mathematical procedure capable to determine the width of the so-called reaction zone following the shock, as well as the Chapman-Jouguet velocity which corresponds to the minimum value of the shock speed such that the existence of the detonation itself is assured. Accordingly, in the present paper, the kinetic modeling of the ZND detonation wave [3] is described for a gas mixture with reversible bimolecular reaction. The structure of the steady solution is represented through the number density profiles and Hugoniot diagrams; the wave thickness behaviour is also shown for different overdrive degrees. On the other hand, the linear stability of the steady detonation constitutes a widely investigated topic after the studies of Erpenbeck [4] who used, for this problem, a Laplace transform method. Further contributions to the evaluation of linear stability spectra are based on the normal mode approach combined with a numerical shooting algorithm, first developed in [5]. An extensive overview of the current state of detonation stability in literature can be found in the review paper [6] by Stewart and Kasimov. Most research works deal with a one-step Arrhenius reaction, whereas only few are related to chain branching chemical reactions [7]. According to the hydrodynamic formulation of paper [5], the modeling here proposed consists in the linearization of the Euler equations and Rankine-Hugoniot conditions through a normal mode expansion about the known steady detonation solution which is recalled in the first part of this paper. The linear stability of the detonation wave is then studied in terms of an initial value problem, providing the evolution of the complex disturbances of the steady state variables due to a small time-dependent perturbation of the shock front position. The Rankine-Hugoniot conditions constitute the initial data at the von Neuman state. A dispersion relation at the end of the reaction zone, such that no acoustic waves propagate from the rear boundary towards the shock to interfere with its steady character, is imposed in order to assure the determinacy of the problem. This closure condition, which expresses the compatibility constraint introduced in Ref. [8] by Buckmaster and Ludford as an acoustic radiation condition, recovers the form discussed in paper [9].

The initial value problem has been numerically treated and the evolution of the complex disturbances in the reaction zone has been obtained for correct growth rate and perturbation frequency. For an elementary reaction of the hydrogen-oxygen system [10], some results concerning the instability behavior are given through the time evolution of the state vector perturbation in the reaction zone. Such results are in good agreement with the main features of the stability analysis known in literature.

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STEADY PROPAGATION OF DETONATION WAVES

Consider a mixture of four gases, of molecular masses m_1, m_2, m_3 and m_4 respectively, with $m_1+m_2=m_3+m_4$, undergoing the reversible bimolecular chemical reaction

$$A_1 + A_2 \rightleftharpoons A_3 + A_4 . \tag{1}$$

A set of Boltzmann-like kinetic equations for such a gas mixture has been derived in paper [1]. Again in that paper a suitable closure at Euler level has been obtained, providing the following set of dimensional hydrodynamic equations for the field variables $\mathbf{u}=(n_1,n_2,n_3,n_4,u,T)^{\mathsf{T}}$

$$\frac{\partial n_i}{\partial t}+\frac{\partial}{\partial x}(n_i u)=s_i S, \quad i=1,\dots,4, \qquad \frac{\partial}{\partial t}(\rho u)+\frac{\partial}{\partial x}(\rho u^2+n\kappa T)=0$$
$$\frac{\partial}{\partial t}(\rho u^2+3n\kappa T)+\frac{\partial}{\partial x}\big[u(\rho u^2+5n\kappa T)\big]=-2ES, \tag{2}$$

where $n=\sum_{i=1}^{4} n_i$, $\rho=\sum_{i=1}^{4} m_i n_i$, $s_i=1$ for $i=1,2$ and $s_i=-1$ for $i=3,4$. Moreover S is the reaction rate [11] that for the reversible chemical reaction (1) is given by

$$S(n_1,n_2,n_3,n_4,T)=4\pi\eta\,\frac{\mu_{34}}{\kappa T}\left[n_3n_4-n_1n_2\Big(\frac{\mu_{34}}{\mu_{12}}\Big)^{3/2}\exp\Big(\frac{-E}{\kappa T}\Big)\right]$$
$$\times\Big\{\frac{\chi}{\pi}\sqrt{\frac{2\pi\kappa T}{\mu_{34}}}\exp\Big[-\frac{\mu_{34}\chi^2}{2\kappa T}\Big]+\Big(\frac{\kappa T}{\mu_{34}}-\chi^2\Big)\Big[1-\mathrm{erf}\Big(\sqrt{\frac{\mu_{34}}{2\kappa T}}\chi\Big)\Big]\Big\}. \tag{3}$$

Above, η is a scalar factor related to chemical collision frequency, χ the exothermic threshold velocity, κ the Boltzmann constant, μ_{ij} the reduced masses and E the activation energy defined by

$$\mu_{ij}=m_im_j/(m_i+m_j), \qquad E=E_3+E_4-E_1-E_2>0,$$

E_i being the bond energies of chemical links and $E>0$ implying that $A_3+A_4\to A_1+A_2$ is exothermic. Under the transformation $z=x-\mathscr{D}t$, where $\mathscr{D}$ denotes the constant speed of a steady detonation wave traveling in the positive $x-$direction towards the quiescent mixture, a re-arrangement of Eqs. (2) leads to the explicit form

$$\frac{dn_i}{dz}=\frac{S}{\mathscr{D}-u}\left[\frac{2En_i}{5\kappa nT-3\rho(\mathscr{D}-u)^2}-s_i\right], \qquad \frac{du}{dz}=\frac{2ES}{5\kappa nT-3\rho(\mathscr{D}-u)^2}$$
$$\frac{dT}{dz}=\frac{2ES[\rho(\mathscr{D}-u)^2-\kappa nT]}{\kappa n(\mathscr{D}-u)[5\kappa nT-3\rho(\mathscr{D}-u)^2]}. \tag{4}$$

A formal integration of Eqs. (4) from the initial state $\mathbf{u}_0=(n_{10},n_{20},n_{30},n_{40},u_0,T_0)^{\mathsf{T}}$ to the von Neumann state leads to the Rankine–Hugoniot conditions, providing the unique jump solution at the von Neumann state, namely

$$n_{i\mathrm{VN}}=\frac{\mathscr{D}-u_0}{\mathscr{D}-u_{\mathrm{VN}}}\,n_{i0}, \quad i=1,\dots,4 \qquad T_{\mathrm{VN}}=\frac{1}{\kappa}\left[\frac{\rho_0(u_{\mathrm{VN}}-u_0)}{n_0}+\frac{\kappa T_0}{\mathscr{D}-u_0}\right](\mathscr{D}-u_{\mathrm{VN}}),$$
$$u_{\mathrm{VN}}=\frac{3}{4}\mathscr{D}+\frac{1}{4}u_0-\frac{5}{4}\frac{n_0\kappa T_0}{\rho_0(\mathscr{D}-u_0)}. \tag{5}$$

Solving Eqs. (4) with initial data (5), the state vector $\mathbf{u}=(n_1,n_2,n_3,n_4,u,T)^{\mathsf{T}}$ can be determined inside the reaction zone. Chemical equilibrium is reached when all the r.h.s of Eqs. (4) vanish, so that the value $z=z_F$, where such a condition occurs, defines the equilibrium final state of the reaction zone. Since Eqs. (2) express the conservation of total mass, momentum and energy of the system, it is possible to obtain in the (V,p)-plane ($V=1/\rho$ and $p=n\kappa T$) the sheaf of Rayleigh lines and the family of Hugoniot curves [2]

$$p=p_0-\rho_0^2(\mathscr{D}-u_0)^2(V-V_0), \tag{6}$$

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$$\left(V-\frac{V_0}{4}\right)\left(p+\frac{p_0}{4}\right) \;=\; \frac{q}{2}+\frac{15}{16}V_0p_0, \tag{7}$$

where $q=\dfrac{\mathscr{E}_{ch0}}{\rho_0}-\dfrac{\mathscr{E}_{ch}}{\rho}$ and $\mathscr{E}_{ch}=\sum_{i=1}^{4}E_in_i$. The Hugoniot curves provide all the detonation states in the (V,p)-plane, while the intersection of these curves with the Rayleigh line (see Fig.1) allows to find the actual solution to the detonation problem for the selected value of $\mathscr{D}$. This set of all possible solutions has always a lower bound for the so called Chapman-Jouguet velocity, $\mathscr{D}_j$, which is reached when the Rayleigh line is tangent to the Hugoniot curve computed for $z=z_F$ (see right picture of Fig.1). The wave thickness versus overdrive degree $f=(\mathscr{D}/\mathscr{D}_j)^2$ is shown in Fig. 2, left picture, and detonation profiles for different values of $\mathscr{D}$ are represented in Fig. 2, right picture, and Fig. 3. The reference data for Figs. 1-3 are the following $m_1=0.018$, $m_2=0.001$, $m_3=0.0017$, $m_4=0.002$, $\beta=10^7$, $\chi=6851$, $E=63311$.

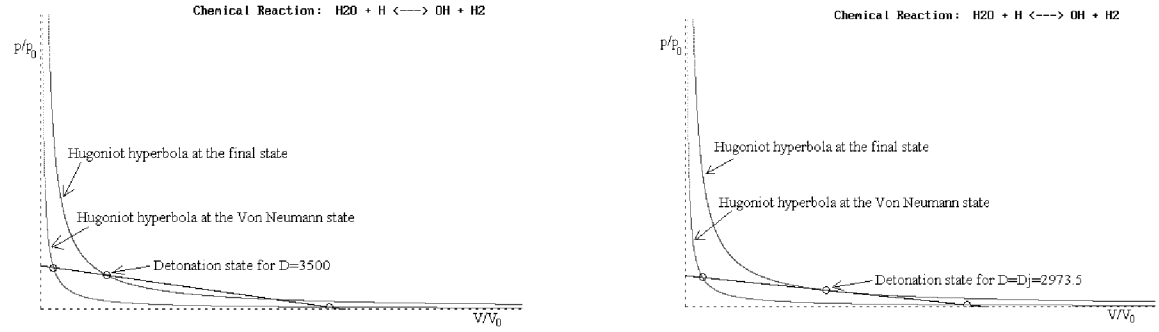

FIGURE 1. Steady solution: Hugoniot diagram with Rayleigh line for $f=1.3854$ (left) and $f=1$ (right).

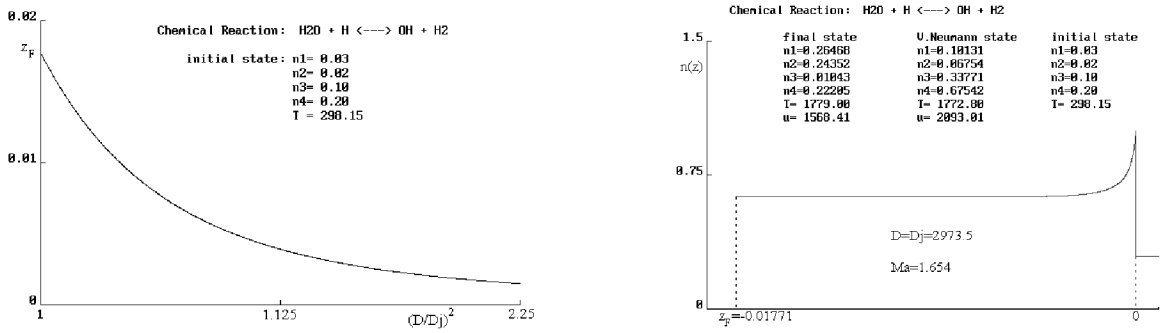

FIGURE 2. Steady solution: wave thickness z_F versus overdrive degree (left) and number density profile for $f=1$ (right).

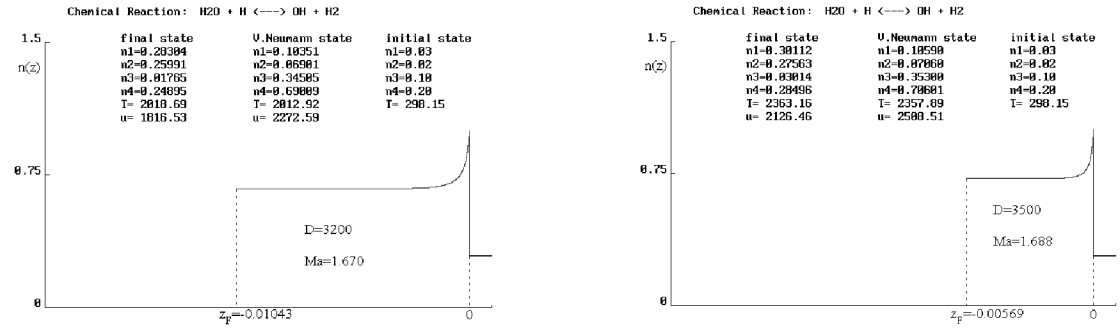

FIGURE 3. Steady detonation profiles: number density for $f=1.1581$ (left) and $f=1.3854$ (right).

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LINEAR STABILITY PROBLEM

The detonation stability is classically studied assuming that a small rear boundary perturbation, instantaneously assigned, induces a distortion on the steady planar shock wave, whereas subsequent rear oscillations do not affect the shock wave [5]. Therefore the shock distortion interferes with the steady character of the state variables in the reaction zone so that the detonation solution does not admit anymore a steady representation. The stability problem consists then in studying the time and space evolution of the state variables perturbations in the reaction zone. An extensive treatment of linear stability within the kinetic frame is developed in a forthcoming paper [12], in the context of the full Boltzmann equation extended to a reversible bimolecular reaction. The starting point consists in the reactive Euler equations of the kinetic model written in their non-steady formulation, together with initial data at the von Neumann state specified by the related Rankine-Hugoniot conditions. In order to deduce the linear stability equations which define the evolution of the responses of the system to the above mentioned perturbations, the governing equations are first re-written in the perturbed shock frame and afterwards linearized through a normal mode expansion. In the new frame, the *wave coordinate* is introduced as $x-[\mathscr{D}t+\psi^{\ell}(t)]$, with $\psi^{\ell}(t)$ the perturbation of the shock position in the laboratory frame. The time derivative becomes then $\partial/\partial t-(\mathscr{D}+d\psi^{\ell}/dt)\,\partial/\partial x$ and the particle velocity is referred to the perturbed shock frame as well, $u=u^*-\mathscr{D}-d\psi^{\ell}/dt$. According to Refs. [5, 7], the distortion of the shock position and perturbation of the state variables are assumed with an exponential time dependence. The normal mode approach proceeds through the expansions around the steady solution, $\mathbf{z}^*=\left[\rho_1^*\,\rho_2^*\,\rho_3^*\,\rho_4^*\,u^*\,p^*\right]^{\mathrm{T}}$, whose components $\rho_i^*=m_i n_i^*$, u^* and $p^*=n^*kT^*$ are determined from the solution to the system (4) and (5). The following decompositions are assumed for the state vector $\mathbf{z}$,

$$\mathbf{z}(x,t)=\mathbf{z}^*(x)+\exp(at)\,\overline{\mathbf{z}}(x),\quad \psi^{\ell}(t)=\overline{\psi}\exp(at),\quad \text{with}\quad a,\overline{\psi}\in\mathbb{C}, \tag{8}$$

where $\mathbf{z}=\left[\rho_1\,\rho_2\,\rho_3\,\rho_4\,u\,p\right]^{\mathrm{T}}$. In Eqs. (8), the overline refers to the unknown perturbations, $\mathrm{Re}\,a=\alpha$ denotes the disturbance growth rate, $\mathrm{Im}\,a=\beta$ the disturbance frequency and $\overline{\psi}$ the perturbation amplitude of the shock front position. The linearization process of the governing equations (2) and (5) through expansions (8) leads to the stability equations in the unknown complex disturbances $\overline{\mathbf{z}}$ and $\overline{\psi}$. The presence of the further unknown $\overline{\psi}$ can be avoided through the renormalization $\overline{\zeta}=\overline{\mathbf{z}}/\overline{\psi}$, so that the stability equations become

$$(A^*-\mathscr{D}I)\,\frac{d\overline{\zeta}}{dx}+(aI+C^*)\,\overline{\zeta}-a\mathbf{b}^*=0 \tag{9}$$

with initial conditions assigned at the von Neuman state,

$$\overline{\zeta}_{\mathrm{VN}}=a(A^*_{\mathrm{VN}}-\mathscr{D}I)^{-1}\,(K^*_{\mathrm{VN}}\mathbf{z}^*_{\mathrm{VN}}-\mathbf{z}_0)\,. \tag{10}$$

The starred quantities refer to the steady solution and $\mathbf{z}_0$ is the initial state vector defining the unreacted fresh mixture. In Eqs. (9,10) $\mathbf{b}^*=d\mathbf{z}^*/dx$ and A^*, C^*, K^*_{VN} are 6×6-matrices defined in paper [12], whose non-zero components are

$$A^*_{ii}=u^*,\ i=1,\dots,6,\qquad A^*_{i5}=\rho_i^*,\ i=1,\dots,4,\qquad A^*_{56}=\frac{1}{\rho^*}\qquad A^*_{65}=\frac{5p^*}{3},$$

$$C^*_{ij}=\delta_{ij}\frac{du^*}{dx}+s_i m_i S^*\rho_h^*\Theta,\ i=1,\dots,4,\ j=1,2,\quad C^*_{ij}=\delta_{ij}\frac{du^*}{dx}-s_i m_i S^*\rho_h^*,\ i=1,\dots,4,\ j=3,4,\quad C^*_{66}=\frac{5}{3}\frac{du^*}{dx},$$

$$C^*_{5j}=\frac{u^*-\mathscr{D}}{\rho^*}\frac{du^*}{dx},\ j=1,\dots,4,\quad C^*_{i5}=\frac{dz_i^*}{dx},\ i=1,\dots,6,\quad C^*_{6j}=\frac{2E}{3}S^*\rho_h^*\Theta,\ j=1,2,\quad C^*_{6j}=-\frac{2E}{3}S^*\rho_h^*,\ j=3,4,$$

$$K^*_{ii}=1,\ i=1,2,3,4,6,\quad K^*_{55}=\sum_i\rho^*_{i0}/\sum_i\rho^*_{iVN},\quad K^*_{65}=-\frac{1}{3}\sum_i\rho^*_{i0}u^*_{VN},$$

where $\Theta=\left(\dfrac{m_3m_4}{m_1m_2}\right)^{5/2}\exp\left(-\dfrac{n^*E}{p^*}\right)$ and S^* is obtained from Eq. (3) with reference to the steady state vector $\mathbf{z}^*$. Moreover δ_{ij} is the Kronecker symbol and the index h is such that $h=2,1,4,3$ when $j=1,2,3,4$, respectively.

The initial value problem (9) and (10), for a given set of chemical and kinetic parameters, provides the x-evolution of the complex disturbances $\overline{\zeta}$ in the reaction zone, in dependence on the complex parameter a, which is not known *a priori*. The determinacy of the stability problem is assured by a compatibility constraint which has the physical

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meaning that no acoustic waves, traveling far away from the reaction zone towards the shock, reach the equilibrium final state. As widely discussed in paper [9] this closure condition can be regarded as the radiation condition variously employed in many research works on the linear detonation stability. Accordingly, in the present paper such condition, which is valid in the equilibrium final state at the end of the reaction zone, $x = x_{\rm F}$, can be written in the form

$$H(a) = 0 \quad \text{for} \quad x = x_{\rm F}, \qquad \text{with} \quad H(a) = \overline{\zeta}_5 + a + \frac{1}{\gamma \rho^*_{\rm F} c^*_{\rm F}} \overline{\zeta}_6 , \tag{11}$$

since the shock travels from the left towards the right and the particle velocity is referred to the perturbed shock frame. In the closure Eq. (11), $\overline{\zeta}_5$ and $\overline{\zeta}_6$ denote the renormalized disturbances of the particle velocity and pressure, $c^*_{\rm F}$ is the isentropic sound speed and γ the ratio of specific heats.

The stability problem formulated by means of Eqs. (9), (10) and (11) has been studied adopting the following strategy, with the aim of characterizing the influence of the perturbations on the steady solution.

- First, for several values of a in a bounded domain of the right complex half-plane, the stability equations (9) have been integrated with initial conditions (10), by means of a fourth order Runge-Kutta routine.
- Afterwards, the entire solution obtained for each trial value of a has been specialized for $x = x_{\rm F}$ in order to compute the residual function $|H(a)|$, defined in the compatibility Eq. (11).
- Only the solutions $\overline{\zeta}$ to ODEs (9) obtained for those values of a for which $|H(a)|$ results to be close to zero, within an acceptable tolerance, are taken into account.

The stability solutions selected as above have been used to evaluate the influence of the perturbations induced by the shock distortion on the steady detonation wave through the decomposition

$$\mathbf{z}(x,t) = \mathbf{z}^*(x) + \exp(at)\,\overline{\zeta}(x)\,. \tag{12}$$

Expressions (12) are obtained from Eqs. (8) after a normalization to 1 of the perturbation amplitude of the shock position, i.e. $\overline{\psi} = 1$. In particular, the influence of the real part of the disturbances on the steady detonation solution $\mathbf{z}^*(x)$ has been considered, that is

$$\mathrm{Re}\left[\exp(at)\,\overline{\zeta}(x)\right] = e^{\alpha t}\left[\sigma(x)\cos\beta t - \tau(x)\sin\beta t\right] \tag{13}$$

where $\sigma = \mathrm{Re}\overline{\zeta}$ and $\tau = \mathrm{Im}\overline{\zeta}$ are the perturbation vectors obtained performing the steps outlined just above.

For the steady detonation solution described in the previous section for overdrive degree $f = 1.3854$, the complex disturbances $\overline{\zeta}$ have been numerically evaluated for disturbance frequency $\beta = 0.01$ and growth rate $\alpha = 0.01$, corresponding to an unstable detonation wave, and for growth rate $\alpha = -0.01$, corresponding to a stable detonation wave. Figures 4 and 5 *(a)* show the increasing behaviour in time of the disturbance amplitudes for different states within the reaction zone, namely the von Neuman state (VN), an intermediate state (I) and the equilibrium final state (F), recovering an instability picture. Left frame of Fig. 4 refers to the disturbance of total mass density, $\overline{\rho} = \overline{\rho}_1 + \cdots + \overline{\rho}_4$, versus time, the right one refers to the disturbance $\overline{p}$ and Fig. 5 *(a)* to the disturbance $\overline{u}$. The pictures reveal that the amplitudes of the disturbances $\overline{\rho}$ are greater at the von Neuman state and smaller at the final state, whereas the opposite occurs for $\overline{p}$ and $\overline{u}$. Conversely, the time decay of the amplitudes of all disturbances has been observed for $\alpha = -0.01$, confirming the stability behaviour. In particular, the decay of $\overline{\rho}$ is represented in Fig. 5 *(b)*.

Another significant feature of the proposed linear stability analysis concerns the existence of perturbation modes which are more unstable for decreasing values of perturbation frequency β, at given positive growth rate.

In conclusion, the kinetic modeling of the linear stability proposed in the last section of the present paper allows to reproduce the main effects induced by the shock distortion on the steady detonation wave. Thus it seems promising to develop a more complete hydrodynamic stability analysis in order to go deeper on the effects due to the heat release and activation energy of the chemical reaction as well to bi-dimensional disturbances [6] induced by a shock distortion of type $\psi^{\ell}(y,t)$.

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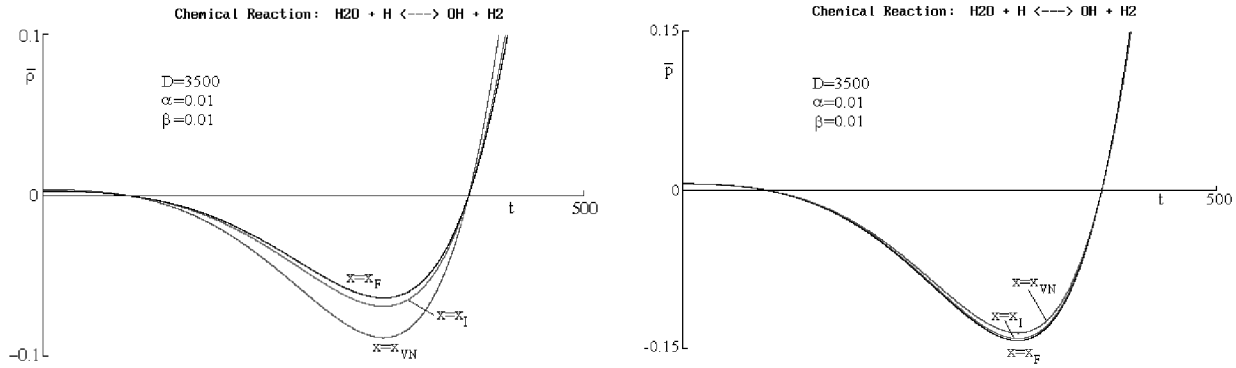

FIGURE 4. Instability behaviour for growth rate $\mathrm{Re}\, a = 0.01$: time evolution of the disturbance amplitudes $\overline{\rho}$ *(left)* and $\overline{\rho}$ *(right)*, at von Neuman ($x = x_{VN}$), intermediate ($x = x_I$) and equilibrium final state ($x = x_F$).

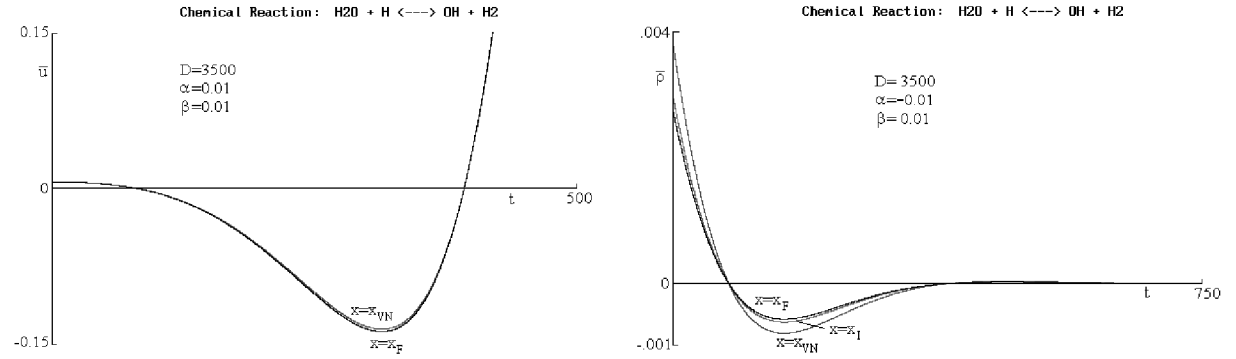

FIGURE 5. *(a)* Instability behaviour for $\mathrm{Re}\, a = 0.01$: time evolution of the disturbance amplitudes $\overline{u}$ at von Neuman ($x = x_{VN}$) and final state ($x = x_F$). *(b)* Stability behaviour for $\mathrm{Re}\, a = -0.01$: time decay of the disturbance amplitudes for $\overline{\rho}$.

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