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# Stability and nonlinear dynamics of one-dimensional overdriven detonations in gases

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The purpose of this analytical work is twofold: first, to clarify the physical mechanisms triggering the one-dimensional instabilities of plane detonations in gases; secondly to provide a nonlinear description of the longitudinal dynamics valid even far from the bifurcation. The fluctuations of the rate of heat release result from the temperature fluctuations of the shocked gas with a time delay introduced by the propagation of entropy waves. The motion of the shock is governed by a mass conservation resulting from the gas expansion across the reaction zone whose position fluctuates relative to the inert shock. The effects of longitudinal acoustic waves are quite negligible in piston-supported detonations at high overdrives with a small difference of specific heats. This limit leads to a useful quasi-isobaric approximation for enlightening the basic mechanism of galloping detonations. Strong nonlinear effects, free from the spurious singularities of the square-wave model, are picked up by considering two different temperature sensitivities of the overall reaction rate: one governing the induction length, another one the thickness of the exothermic zone. A nonlinear integral equation for the longitudinal dynamics of overdriven detonations is obtained as an asymptotic solution of the reactive Euler equations. The analysis uses a distinguished limit based on an infinitely large temperature sensitivity of the induction kinetics and a small difference of specific heats. Comparisons with numerical calculations show a satisfactory agreement even outside the limits of validity of the asymptotic solution.

## 1. Introduction

The evidence that gaseous detonation waves might prefer to travel with a locally three-dimensional and unsteady configuration has been established experimentally for many years (see for a review: Fickett & Davis 1979; Strehlow 1985; Williams 1985). The boundaries of the cells developing on detonation fronts are Mach stems propagating in the transverse direction across the shocked gas at approximately acoustic velocity, so that the pattern changes continuously with time. Smoked-foil records of detonations in tubes exhibit diamond-shaped patterns which have been reproduced by two-dimensional direct numerical simulations (Oran & Boris 1987; Bourlioux & Majda 1992). From the theoretical side, the most complete treatment of stability of detonation waves is that of Erpenbeck (1964). Solving the initial-value problem by Laplace transformation, he obtained the stability limits numerically but could not expose the underlying physical mechanisms. Abouseif & Toong's statement (1982), 'Many attempts to unravel the physical mechanism triggering the detonation instability have met with a modicum of success', still holds. A coupling of transverse acoustic waves to a longitudinal oscillatory instability could be the key point (Abouseif & Toong 1986).

One-dimensional oscillations, called galloping detonations, were predicted by direct numerical simulations of Fickett & Wood (1966) and observed by shooting a blunt body projectile into reactive mixtures (Alpert & Toong 1972; Lehr 1972; see also the preliminary results of 1962 at NBS which are reported in the *9th Symposium on Combustion*, p. 476). The corresponding Hopf bifurcation has been described by numerical analysis of the linearized equations (Erpenbeck 1964; Lee & Stewart 1990). Erpenbeck (1967) was the first to develop a weakly nonlinear theory. The general bifurcation formalism leading to the Landau–Stuart equation has been adapted to this problem by Bourlioux, Majda & Roytburd (1991). Despite all the information that can be extracted from these formal analyses, they fail to provide enough physical insights into the problem for its modelling. The purpose of the present analytical work is twofold: first, to clarify the physical mechanisms governing the one-dimensional instabilities of planar detonations; secondly, to provide a description of the nonlinear dynamics valid even far from the bifurcation. Moreover, the asymptotic solution of the reactive Euler equations presented in this paper in one-dimensional geometry is a prerequisite for tackling the multi-dimensional and unsteady structures of detonations.

The governing equations written in terms of a mass-weighted coordinate are presented in §2. Section 3 is devoted to simplified models which provide the physical insights into the problem. Section 4 is concerned with a rational approximate solution to the reactive Euler equations. A nonlinear integral equation for the dynamics of overdriven detonations is obtained by an asymptotic analysis. The results are discussed in §5. Conclusions and perspectives are presented in §6.

Since the first works by Shchelkin (1959) and Zaidel (1961), there has been much discussion in the literature of the square-wave model (see Fickett & Davis 1979 for a review). This model which corresponds to the singular zero limit of the ratio of the reaction time to the induction time, proved to be very useful in describing nonlinear phenomena in quasi-steady regimes of propagation (Zeldovich 1940; He & Clavin 1992, 1994). However, the detonation dynamics obtained from this model is singular. The one-dimensional stability analysis of Erpenbeck (1963) yields an infinite spectrum of discrete unstable modes with unbounded linear growth rates increasing with frequency. This differs drastically from the stable modes obtained at high frequency by the numerical results of a smoothly distributed heat release. The square-wave model leads to a differential-difference equation for the shock velocity which is of the advanced type: the velocity of the leading shock at the current time depends on both the shock velocity and the shock acceleration at an earlier time (Fickett 1985*b*). Solutions of such equations develop singularities after a finite time. However, the physical mechanisms involved in this singular dynamics are worth investigating: we show in §§3.2 and 3.3 that the conservation of mass between the leading shock and the piston is responsible for the one-dimensional instability of piston-supported detonations with a large degree of overdrive. Acoustic signals play a minor role. The sharp time delay due to propagation of entropy waves is at the origin of the singular dynamics. A simple example illustrating the drastic effect of a distributed rate of heat release on the spectrum of the linear modes is presented in §3.4. The singular character of the dynamics is suppressed by averaging the time delay over space.

A ratio of specific heat close to unity is an approximation which simplifies greatly the nonlinear analysis of the reactive Euler equations (Blythe & Crighton 1989): in the entropy waves, the temperature fluctuations are mainly due to the variations of the heat release rate; the entropy waves are no longer coupled to the pressure field and the unsteady distributions of temperature and reaction rate are governed by the temperature of the shocked gas only (see §4.1). When the temperature sensitivity of the induction kinetics is large, strong nonlinear effects are picked up even with small fluctuations of the shock velocity (see §4.2). As the square-wave model results from the Arrhenius law in the limit of infinitely large activation, a more sophisticated asymptotic expansion must be used to avoid spurious singularities. Two different temperature sensitivities are introduced; one governing the induction length and another one the distribution of heat release rate. Only the first one is considered as a large parameter. Such approximations are accurate for gaseous detonations. Finally, the additional assumption of a high degree of overdrive is used for exhibiting the essential role of mass conservation within a quasi-isobaric approximation. In this limiting regime of propagation the longitudinal acoustic waves have a negligible effect on the longitudinal oscillatory instability. A nonlinear integral equation is obtained in §4.3 by an asymptotic analysis of the Euler reactive equations in a distinguished limit associated with the above mentioned approximations. The linear version of this equation bridges the gap between the unphysical character of the singular instability exhibited by the square-wave model and the stable solutions observed with a sufficiently smooth structure. The nonlinear dynamics obtained by this equation is compared with direct numerical simulations of one-dimensional detonations. Satisfactory agreement is obtained even outside the limits of validity of the analysis. The ways to extend these results to low overdrives and to a multi-dimensional geometry are briefly outlined.

## 2. Governing equations

The governing equations in a one-dimensional, inviscid, chemically reacting gas flow may be written as follows:

$$\frac{\partial \rho}{\partial t}+\frac{\partial(\rho u)}{\partial x}=0, \qquad (2.1\,a)$$

$$\frac{\partial(\rho u)}{\partial t}+\frac{\partial(\rho u^2+p)}{\partial x}=0, \qquad (2.1\,b)$$

$$C_p\left(\frac{\partial T}{\partial t}+u\frac{\partial T}{\partial x}\right)-\frac{1}{\rho}\left(\frac{\partial p}{\partial t}+u\frac{\partial p}{\partial x}\right)=QW, \qquad (2.1\,c)$$

$$\frac{\partial(\rho y)}{\partial t}+\frac{\partial(\rho u y)}{\partial x}=\rho W, \qquad (2.1\,d)$$

with

$$p=(C_p-C_v)\rho T, \qquad (2.1\,e)$$

for an ideal gas. Variables are as follows: $t$ is the time, $x$ the distance, $u$ the gas velocity, $p$ the pressure, $\rho$ the mass density, $T$ the temperature, $y$ the progress variable ($y=0$ in the initial reactive mixture, $y=1$ in the burned gases), $C_p$ and $C_v$ the specific heats, $Q$ the heat of reaction and $W$ the reaction rate per unit mass. The simplest model of chemical reaction corresponds to a reactant which is converted to a product by a one-step irreversible exothermic reaction. In such a case, $(1-y)$ is the reactant mass fraction and the reaction rate $W(y,T)$ is a function of $y$ and $T$. The most popular kinetic model in gaseous combustion is the Arrhenius law,

$$W=t_{col}^{-1}B(1-y)^n\exp-(E/RT), \qquad (2.1\,f)$$

where $t_{col}$ is the elastic collision time, $B$ a prefactor and $n$ the order of the reaction. A large activation energy, $E/RT_N \gg 1$, ensures the validity of the ZND detonation structure in which an inert shock wave is followed by a reaction zone; $T_N$ is the temperature of the shocked gas just downstream the shock (Neumann spike).

### 2.1. *Mass-weighted coordinate*

Let $x_s(t)$ be the path of the leading shock. As in the study of vibratory instabilities of planar flames by Clavin, Pelce & He (1990), it is convenient to introduce a system of reduced coordinates constituted by a mass-weighted dimensionless distance relative to the moving shock and a reduced time,

$$\xi = \frac{1}{t_{ref}\rho_u D_0}\int_{x_s(t)}^{x} \rho \,\mathrm{d}x, \quad \tau = \frac{t}{t_{ref}}, \tag{2.2a}$$

$D_0$ is the shock velocity of the steady detonation wave propagating into the fresh gas mixture initially at rest with a density $\rho_u$. The timescale $t_{ref}$ is arbitrarily chosen, for example as the induction time,

$$t_{ref} = \alpha W^{-1}(y=0, T_{N_o}), \tag{2.2b}$$

where $\alpha$ is a coefficient chosen for convenience and subscript $o$ denotes the steady state. With these reduced coordinates the governing equations are written in the moving frame attached to the leading shock as

$$-\frac{\mathrm{D}}{\mathrm{D}\tau}\left(\frac{\rho_u}{\rho}\right) + \frac{\partial}{\partial \xi}\left(\frac{v}{D_o}\right) = 0, \tag{2.3a}$$

$$\frac{\mathrm{D}}{\mathrm{D}\tau}v + \frac{1}{\rho_u D_o}\frac{\partial p}{\partial \xi} = 0, \tag{2.3b}$$

$$\frac{1}{T}\frac{\mathrm{D}}{\mathrm{D}\tau}T - \left(\frac{\gamma-1}{\gamma}\right)\frac{1}{p}\frac{\mathrm{D}}{\mathrm{D}\tau}p = \frac{Q}{C_p T}w(y,T) \quad \text{or} \quad \frac{1}{\gamma}\frac{1}{p}\frac{\mathrm{D}}{\mathrm{D}\tau}p - \frac{1}{\rho}\frac{\mathrm{D}}{\mathrm{D}\tau}\rho = \frac{Q}{C_p T}w, \tag{2.3c}$$

$$\frac{\mathrm{D}}{\mathrm{D}\tau}y = w(y,T), \tag{2.3d}$$

where $w \equiv Wt_{ref}$ is the reduced reaction rate, $v \equiv D_0 + u$ the gas velocity relative to the shock of the steady detonation and $\gamma = C_p/C_v$ the ratio of specific heat. $\mathrm{D}/\mathrm{D}\tau$ denotes the substantive derivative in reduced coordinates (2.2),

$$\frac{\mathrm{D}}{\mathrm{D}\tau} \equiv \frac{\partial}{\partial \tau} + m(\tau)\frac{\partial}{\partial \xi}, \tag{2.3e}$$

and

$$m(\tau) \equiv \frac{\rho_N(\tau)}{\rho_0 D_0}[u_N(\tau) + D(\tau)] = \frac{D(\tau)}{D_0} \tag{2.3f}$$

is the reduced mass flux across the leading shock moving with a velocity $D(\tau) \equiv -\mathrm{d}x_s/\mathrm{d}t$. The gas velocity relative to the perturbed shock is $u + D = v - D_o + D$. Subscript $N$ denotes the state at $\xi = 0$, immediately downstream of the shock front of the unsteady detonation (Neumann spike for a steady CJ-detonation). The propagation is in the negative direction, and the gas flow downstream of the shock corresponds to $\xi \geqslant 0$. When $\rho$ is eliminated from (2.3$a$) and (2.3$c$), one gets

$$\frac{1}{\gamma p}\frac{\mathrm{D}}{\mathrm{D}\tau}p + \frac{\rho}{\rho_u}\frac{\partial}{\partial \xi}\left(\frac{v}{D_o}\right) = \frac{Q}{C_p T}w. \tag{2.4}$$

Then, by combining (2.4) with (2.3*b*), the characteristic equations for a reacting gas are obtained in terms of the reduced mass-weighted coordinates:

$$\frac{1}{\gamma p}\frac{\mathrm{D}^{\pm}}{\mathrm{D}\tau}p \pm \frac{1}{a}\frac{\mathrm{D}^{\pm}}{\mathrm{D}\tau}v = \frac{Q}{C_p T}w(y, T), \tag{2.5a–b}$$

with

$$\frac{\mathrm{D}^{\pm}}{\mathrm{D}\tau} \equiv \frac{\partial}{\partial \tau} + \left[m(\tau) \pm \frac{\rho a}{\rho_u D_0}\right]\frac{\partial}{\partial \xi}, \tag{2.5c}$$

where $\rho(p, T)$ is given by (2.1*e*) and where the frozen sound speed $a(T)$ is defined by

$$a^2 \equiv \gamma \frac{p}{\rho} = \gamma (C_p - C_v)\, T. \tag{2.5d}$$

The motion of the leading shock, $m(\tau)$, is obtained from the solution of hyperbolic equations (2.3*c, d*) and (2.3*a, b*) or (2.5*a, b*) satisfying both the Rankine–Hugoniot jump conditions across the inert shock at $\xi = 0$ and an additional boundary condition in the burned gas at $\xi \rightarrow +\infty$ (see below).

### 2.2. *Boundary conditions*

When the state of the fresh mixture (labelled by subscript $u$) in which the detonation propagates, is uniform and constant, the values of all the variables ($p_N$, $v_N$, $T_N$, $y_N$) at $\xi = 0$ are expressed in terms of $m(\tau)$ by using the Rankine–Hugoniot conditions,

$\xi = 0$:

$$\frac{p_N}{p_u} = \left(\frac{2\gamma}{\gamma+1}\right)M^2 - \left(\frac{\gamma-1}{\gamma+1}\right) \approx \left(\frac{2\gamma}{\gamma+1}\right)M^2, \tag{2.6a}$$

$$\frac{(v_N + D - D_0)}{D} = \frac{\rho_u}{\rho_N} = \left(\frac{\gamma-1}{\gamma+1}\right) + \left(\frac{2}{\gamma+1}\right)\frac{1}{M^2}, \tag{2.6b}$$

$$M_N^2 = \frac{2+(\gamma-1)M^2}{2\gamma M^2 - (\gamma-1)} \approx \frac{2+(\gamma-1)M^2}{2\gamma M^2}, \quad \frac{T_N}{T_u} = \frac{\{2\gamma M^2 - (\gamma-1)\}\{(\gamma-1)M^2+2\}}{(\gamma+1)^2 M^2}$$

$$\approx \frac{2\gamma}{(\gamma+1)^2}\{(\gamma-1)M^2+2\}, \tag{2.6c}$$

$$y_N = 0 \quad \text{(inert shock)}, \tag{2.6d}$$

where $M$ is the Mach number of the leading shock,

$$M \equiv D/a_u = m(\tau)\, M_o, \tag{2.6e}$$

and where $M_o$ is the Mach number of the steady solution $M_o \equiv D_o/a_u$. $M_N$ is the local Mach number of the flow immediately downstream of the shock,

$$M_N \equiv \frac{(v_N + D - D_0)}{a_N} < 1, \tag{2.6f}$$

where $a_N \equiv a(T_N)$ is given by (2.5*d*). The approximation $M^2 \gg (\gamma-1)/2\gamma$ with $(\gamma-1)M^2 = O(1)$ which has been used in (2.6), is valid for gaseous detonations. Finally, let us recall the Mach number of a CJ detonation:

$$M_{CJ} = \left[1 + \left(\frac{\gamma+1}{2}\right)\frac{Q}{C_p T_u}\right]^{1/2} + \left[\left(\frac{\gamma+1}{2}\right)\frac{Q}{C_p T_u}\right]^{1/2}$$

so that

$$Q/C_p T_u \gg 1 \Rightarrow M_{CJ}^2 \approx 2(\gamma+1)\, Q/C_p T_u \gg 1. \tag{2.6g}$$

The usual boundary conditions in the burned gases depend on the cases under considerations:

(i) For an overdriven detonation sustained by a moving piston, the gas velocity $v$ is prescribed at the position of the piston. In the limiting case of a piston moving at constant velocity $v_{b_o}$ far from the reaction zone, one has

$$\xi \to +\infty: \quad v = v_{b_o} \Rightarrow \delta v = 0. \tag{2.7a}$$

A steady solution exists only for a subsonic piston velocity, $v_{b_o} \leqslant a_{b_o}$, i.e. $M_{b_o} \leqslant 1$, where subscript $b$ denotes the burned gas state, $M_{b_o} \equiv v_{b_o}/a_{b_o}$ is the Mach number.

(ii) For the same configuration, a different condition, namely an acoustic radiation condition, is also often used in the stability analysis. When each variable $z$ is decomposed as $z = z_o + \delta z$, the acoustic radiation condition is written as

$$\xi \to +\infty: \quad \delta p - \rho_{b_o} a_{b_o} \delta v = 0. \tag{2.7b}$$

This condition applies at the end of the reaction and results from the acoustic solution in the rear region of burned (inert) gases between the end of the reaction and the piston. Its validity rests upon the assumption that the reaction thickness is much smaller than the distance of the piston from the leading shock. According to the linearized form of (2.5$b$) with $w = 0$, the forward characteristic propagating in the burned gases yields

$$\{\partial/\partial\tau + (1 - M_{b_o}^{-1})\,\partial/\partial\xi'\}\,\delta f_{b-} = 0 \quad \text{where} \quad \delta f_{b-} \equiv \delta p - \rho_{b_o} a_{b_o} \delta v, \tag{2.8a}$$

with a general solution of the form

$$\delta f_{b-}(\tau, \xi') = f\left(\tau + \frac{\xi'}{M_{b_o}^{-1} - 1}\right) \quad \text{with} \quad M_{b_o} < 1. \tag{2.8b}$$

The boundedness requirement at the piston ($\xi' = +\infty$) in the unstable case ($\delta f_{b-} \propto \mathrm{e}^{\sigma\tau}$ with $Re\,\sigma > 0$) implies that $\delta f_{b-} = 0$ in all the rear zone. Matching conditions of the thin reaction layer with the large burned gases region yields boundary condition (2.7$b$).

(iii) For a freely propagating detonation followed by a rarefaction wave, a sonic condition must be used in the burned gas region. As for CJ waves, this condition is necessary to protect the shock-reaction complex from quenching by the rarefaction wave. This problem will not be considered here.

The intrinsic longitudinal dynamics of detonations develop on the induction timescale and are not very sensitive to details of the rear boundary condition. This is confirmed by the numerical analysis of Lee & Stewart (1990) showing that the stability limits and the linear spectrum obtained with a piston condition (2.7$a$) or a radiation condition (2.7$b$) are quite similar.

## 3. Physical considerations

Before analysing the one-dimensional dynamics of planar detonations in a systematic way, we present in this section studies of simplified models which are useful for providing the physical background of the perturbation method which is used in §4 for deriving an asymptotic solution of the reactive Euler equations. To begin with, we clarify the mechanism responsible for the singular character of the dynamics of plane detonations put in evidence by Erpenbeck (1963) within the approximation of the square-wave model. The stiffness of this model (discontinuity in the rate of heat release) induces spurious singularities. However, the mechanisms involved are worth investigating because they are limiting forms of those triggering the instability of real detonations. The one-dimensional stability analysis of the square-wave model is briefly revisited in the next section.

### 3.1. *Square-wave model*

In this model all of the heat is released instantaneously after a state-dependent induction time $t_i$. The leading shock is followed by the constant-state induction zone of thickness $l_i$ which is terminated by an infinitely thin exothermic layer located at $x_r$, $l_i \equiv x_r - x_s$, as sketched in figure 1. This model which corresponds to the singular zero limit of the ratio of the reaction time to the induction time, $(t_r/t_i) \to 0$, may be obtained from an Arrhenius law (2.1*f*) in the asymptotic limit $E/RT_{N_o} \to +\infty$. The main results are presented below and details are given in the Appendix. When the Rankine–Hugoniot conditions are used, the linear solutions for both the acoustic and entropy waves propagating across the induction zone express the perturbations of the gas flow at the entrance of the reaction sheet at the current time $\tau$, $\delta p_r(\tau)$, $\delta v_r(\tau)$, $\delta \rho_r(\tau)$, as a linear combination of the fluctuations of the reduced mass flux crossing the leading shock, but at three different instants of time,

$$\delta m(\tau - 1), \quad \delta m(\tau - 1 + \Delta\tau_1), \quad \delta m(\tau - 1 + \Delta\tau_2) \tag{3.1}$$

with three delays: one is the reduced transit time of the entropy waves from the leading shock to the reaction sheet, $1 - \Delta\tau_1$ and $\Delta\tau_2 - 1$ are positive time delays associated with the acoustic signals propagating in the downstream and upstream directions across the induction zone,

$$1 - \Delta\tau_1 \equiv 1/(M_{N_o}^{-1} + 1), \quad \Delta\tau_2 - 1 \equiv 1/(M_{N_o}^{-1} - 1), \tag{3.2}$$

where $M_{N_o} < 1$ is the local Mach number of the flow at the Neumann spike. In the limit $(t_r/t_i) \to 0$, the reaction sheet is in quasi-steady state and responds instantaneously to any perturbations $\delta p_r(\tau)$, $\delta v_r(\tau)$, $\delta \rho_r(\tau)$ which vary with the same characteristic timescale as the induction time, $\Delta\tau = O(1)$. Then the fluctuations of the reduced mass flux crossing the reaction sheet $\delta m_r(\tau)$ may be expressed as a linear combination of $\delta p_r(\tau)$, $\delta v_r(\tau)$ and $\delta \rho_r(\tau)$ yielding

$$\delta m_r(\tau) = c\,\delta m(\tau - 1) + b_1\,\delta m(\tau - 1 + \Delta\tau_1) + b_2\,\delta m(\tau - 1 + \Delta\tau_2). \tag{3.3}$$

The coefficients $c$, $b_1$ and $b_2$ are obtained from the boundary conditions in the burned gases, (2.7*a*) or (2.7*b*), and the jump conditions corresponding to conservation of mass, momentum and total enthalpy across the quasi-steady reaction sheet. Their values are not useful here. Let $\beta$ be a reduced activation energy representing the temperature sensitivity of the induction time, $\beta$ is a large number with a typical value 10. Then kinematic and kinetic considerations presented in the Appendix lead to

$$-\beta(1/T_{N_o})\,\mathrm{d}T_N(\tau - 1)/\mathrm{d}\tau = \delta m_r(\tau) - \delta m(\tau - 1), \tag{3.4}$$

expressing the time derivative of the induction time of a fluid particle reacting at $\tau$ and shocked at $\tau - 1$. All the ingredients are now collected to describe the strong instability experienced by a detonation wave in the limiting case of the square-wave model. According to the Rankine–Hugoniot condition (2.6*c*), $\delta T_N(\tau - 1)/T_{N_o}$ is proportional to $\delta m(\tau - 1)$, then (3.3) and (3.4) lead to an evolution equation for $\delta m(\tau - 1)$ which involves two time delays only. After a shift of origin $(\tau - 1 \to \tau)$ the equation can be written as

$$\mathrm{d}m(\tau)/\mathrm{d}t = C\,\delta m(\tau) + B_1\,\delta m(\tau + \Delta\tau_1) + B_2\,\delta m(\tau + \Delta\tau_2), \tag{3.5}$$

where, according to (3.2), $\Delta\tau_1 > 0$ and $\Delta\tau_2 > 0$ are the difference of transit times

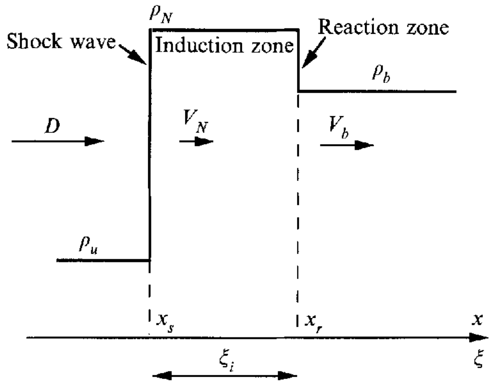


FIGURE 1. Sketch of a square-wave detonation structure.

between the entropy wave and the acoustic signals. Such a difference-differential equation of the advanced type presents singularities after a finite time.

### 3.2. *Physical mechanisms of the oscillatory instability of detonations*

In the approximate analysis developed by Abouseif & Toong (1982) to elucidate the instability mechanisms of less singular models of detonation, the attention was focused on the coupling of the exothermic reaction to the acoustic waves propagating between the inert shock and the end of the exothermic reaction. As for the thermo-acoustic instabilities of combustion in a cavity or of vibratory flames propagating in tubes (Clavin *et al.* 1990; Searby & Rochwerger 1991; Clavin & Sun 1991), amplification of acoustic waves generated by the perturbed heat source was considered as an important mechanism of galloping detonations. A somewhat different understanding arises by noticing that the singular instability of the square-wave model remains even when the acoustic waves are removed from the induction zone in the zero Mach number limit $M_{N_o} \to 0$. According to (2.6*c*), such a small Mach number approximation is valid for a strong shock wave and a small difference of specific heats:

$$(\gamma-1)\,M_o^2 > 1, \quad (\gamma-1) \ll 1 \Rightarrow M_{N_o} \ll 1. \tag{3.6a}$$

According to (3.2), the two time delays of (3.5) are identical in this limit: $\Delta\tau_1 \to 1$, $\Delta\tau_2 \to 1$. Equation (3.5) reduces to a differential equation of the advanced type but with only one time delay; the catastrophic instability is still present,

$$\mathrm{d}m(t)/\mathrm{d}t = C\,\delta m(t) + (B_1+B_2)\,\delta m(t+1). \tag{3.6b}$$

In order to clarify the origin of this instability, consider the piston problem (2.7) sketched in figure 1 in a simplified version in which the density fluctuations are negligible both in the burned gases and in the induction region, $\delta\rho/\rho \ll 1$, but with a density jump across the reaction of order unity and with a fluctuation of position of the reaction sheet relative to the shock of order unity, $\delta l_i/l_i = O(1)$. The time derivative of the mass by unit cross-section between shock and piston is governed by the variation of the mass flux across the shock $\rho_u\,\delta D(t)$. This mass variation may be also expressed in terms of the time derivative of the distance of the reaction sheet from the shock $\mathrm{d}l_i/\mathrm{d}t$. By equating these two expressions, mass conservation yields

$$\delta \mathrm{D}(t) = -\left(\frac{\rho_{N_o}-\rho_{b_o}}{\rho_{b_o}-\rho_{u_o}}\right)\frac{\mathrm{d}}{\mathrm{d}t}l_i(t). \tag{3.6c}$$

Owing to the induction kinetics, the time derivative of the induction length $(1/l_{i_o})\,\mathrm{d}l_i(\tau)/\mathrm{d}\tau$ is related to the time derivative of the temperature fluctuations at the shock but at an early time involving a delay associated with the transit time of the entropy wave $(1/T_{N_o})\,\mathrm{d}T_N(\tau-1)/\mathrm{d}\tau$ (see the Appendix). According to the Rankine–Hugoniot condition (2.6*c*), $(1/T_{N_o})\,\mathrm{d}T_N(\tau-1)/\mathrm{d}\tau$ is proportional to $(1/D_o)\,\mathrm{d}D(\tau-1)/\mathrm{d}\tau$. This yields the same evolution equation as (3.6*b*) with, according to (2.6*b*) and (3.6*a*), $\rho_u/\rho_{N_o} = O(\gamma-1)$ and $C = O(\gamma-1)$, $(B_1+B_2)^{-1} = O(\beta(\gamma-1))$. To summarize, the basic mechanisms involved here are:

(i) the motion of the reaction sheet with a velocity relative to the shock governed by the velocity fluctuations of the leading shock but at an earlier time;

(ii) the transit time of entropy waves from shock to reaction sheet;

(iii) a mass conservation involving the gas expansion across the moving reaction sheet.

The role of entropy waves was clearly identified in the approximate analysis by Abouseif & Toong (1982) but the attention was focused on the compressible effects of acoustic waves. As shown by the comparison between (3.5) and (3.6*b*), the propagation of acoustic signals may influence the final result by modifying the delay, but is not the key mechanism of the oscillatory instability. In the limiting case of piston-supported detonations with a sufficiently large degree of overdrive, the effects of acoustic waves are quite negligible yielding a simple description of galloping detonations which is still relevant, at least qualitatively, outside the domain of validity of this approximation. The discontinuous change of density across the reactive sheet is responsible for the spurious singularities of the square-wave model. For real detonations with a distributed rate of heat release, averaging the time delay over space suppresses the singular character as shown in §3.4. This is more easily described at high overdrives presented below.

### 3.3. *Quasi-isobaric approximation*

A small local Mach number approximation (3.6*a*) is valid in the induction zone only. For piston-supported detonations with a sufficiently high degree of overdrive, the local Mach number is also small throughout the exothermic region. The degree of overdrive $f$ is defined as the square of the ratio of the propagation velocity $D$ to that of the Chapman–Jouguet detonation $D_{CJ}$. Thus, $f \equiv M_o^2/M_{CJ}^2$ where $M_{CJ}$ is given by (2.6*g*). The Mach number in the burned gases $M_{b_o}$ behaves as

$$(\gamma-1)\to 0,\quad f^{-1}\to 0 \Rightarrow M_{b_o}^2 \approx \tfrac{1}{2}\{(\gamma-1)+f^{-1}\}, \tag{3.6d}$$

showing the domain of validity of a uniformly valid approximation of a small local Mach number. Notice that the Euler reactive equations (2.1) are still valid in detonations with a small local Mach number approximation. The relative importance of the molecular transport processes compared to the convective ones is measured by the inverse of a Reynolds number, $\nu/lv_{N_o} \approx M_{N_o}^{-2}t_{col}/t_{reac}$, built with a characteristic diffusion coefficient, $\nu \approx a_{N_o}^2 t_{col}$ and a characteristic length of variation, $l \approx t_{reac}v_{N_o}$, where $t_{reac}$ is a characteristic reaction time. According to (2.1*f*), $t_{col}/t_{reac} \approx \exp(-E_{N_o}/RT_{N_o})$, and the validity of the reactive Euler equations corresponds to an intermediate range of local Mach numbers,

$$\exp-(E_{N_o}/RT_{N_o}) \ll M_{N_o}^2 \ll 1, \tag{3.6e}$$

quite relevant for gaseous detonations.

As shown by (2.5*a, b*) with $w = 0$, the order of magnitude of acoustic-induced isentropic pressure fluctuations is $\delta p/p \approx M_1\,\delta v/v$ where $M_1 \approx v/a$ is the local Mach number. Order of magnitude estimates of pressure fluctuations due to gas velocity variations on the time and lengthscales of the induction period, are according to (2.3$b$), $\delta p/p \approx M_1^2 \delta v/v$. According to (2.6$a$) the pressure fluctuations of the shocked gas are negligible when the perturbations of the detonation velocity are small:

$$\delta p_N/p_N \approx 2\delta D/D \ll 1. \tag{3.6$f$}$$

When (3.6$a$) and (3.6$e,f$) are satisfied, the pressure fluctuations $\delta p/p$ are negligible in (2.4) which reduces to a mass conservation resulting from a quasi-isobaric gas expansion as in flames (see Clavin 1994 for a recent review). Boundary conditions (2.7$a,b$) are identical in this approximation and mass conservation yields

$$\frac{v_b - v_N}{D_o} = \frac{\{(\gamma-1)M_o^2+2\}}{(\gamma+1)M_o^2}\frac{Q}{C_p T_{N_o}}\int_0^\infty w\,\mathrm{d}\xi, \tag{3.7$a$}$$

which is valid for any profile of heat release. Equation (3.7$a$) has to be used with the Rankine–Hugoniot conditions (2.6$b,c$) written in the linear approximation as

$$\frac{\delta v_N}{D_o} \approx -\frac{\delta D}{D_o} \approx -\frac{\{(\gamma-1)M_o^2+2\}}{2(\gamma-1)M_o^2}\frac{\delta T_N}{T_{N_o}}. \tag{3.7$b$}$$

Whenever the combustion laws are such that the distribution of the rate of heat release $w = \Omega(\xi, \tau)$ is expressed in terms of the temperature fluctuations at the shock $\delta T_N/T_{N_o}$ only, (3.7$a,b$) yield the evolution equation of strongly overdriven detonations. According to (2.6$b$) and (3.7$b$), $v_N/D_o \approx (\gamma-1)$ and $\delta v_N/v_N \approx -(\gamma-1)^{-1}\delta D/D$, in such a way that in the limiting case (3.6$a$), strong effects are retained in (3.7$a$) even for small perturbations of the detonation velocity, $\delta D/D = O(\gamma-1)$.

### 3.4. *Effects of a distributed rate of heat release. Comparison with the square-wave model*

In order to illustrate how a distributed rate of heat release changes the detonation dynamics from the singular one, (3.5) or (3.6$b$), let us consider a model which is just the opposite of the square-wave model. Assume that the reactive fluid particle liberates heat with a constant reaction time prescribed by the temperature at which the particle was shocked. In the same notations as (2.1$f$), this yields for $n = 1$,

$$w = \Omega(\xi, \tau) = \frac{t_{ref}}{t_{col}}[1 - y(\xi,\tau)]\exp\left\{-\frac{E}{RT_N(\tau-\xi)}\right\}. \tag{3.8$a$}$$

By choosing the reference time as

$$t_{ref} \equiv t_{col}\exp(-E/RT_{N_o}), \tag{3.8$b$}$$

(3.8$a$) is written as

$$w \approx (1-y)/\lambda_r(\tau-\xi) \quad \text{with} \quad \lambda_r(\tau) \equiv \exp(-\Theta_N(\tau)) \quad \text{and} \quad \Theta_N \equiv \beta\delta T_N/T_{N_o},$$

where

$$\beta \equiv E/RT_{N_o} \gg 1, \quad \Theta_N = O(1). \tag{3.8$c$}$$

According to (3.8$c$), the attention is focused on small perturbations at the leading shock which are amplified by a high temperature sensitivity $\beta \gg 1$: $\delta D/D_o = \delta m = O(1/\beta) \Leftrightarrow \delta T_N/T_{N_o} = O(1/\beta) \Rightarrow \delta w/w = O(1)$. In such a case, $\delta m$ may be neglected in (2.3$d$) to give

$$\frac{\partial}{\partial \tau}y + \frac{\partial}{\partial \xi}y = w, \tag{3.8$d$}$$

with a solution satisfying the boundary condition at $\xi = 0$: $y = 0$ of the form:

$$(1-y) = \exp\left(-\frac{\xi}{\lambda_r(\tau-\xi)}\right), \quad w = \Omega(\xi,\tau) = \frac{1}{\lambda_r(\tau-\xi)}\exp\left(-\frac{\xi}{\lambda_r(\tau-\xi)}\right), \tag{3.8e}$$

to give, in the linear approximation,

$$w = \mathrm{e}^{-\xi} + \delta\Omega(\xi,\tau) \quad \text{with} \quad \delta\Omega(\xi,\tau) = (1-\xi)\,\mathrm{e}^{-\xi}\Theta_N(\tau-\xi). \tag{3.8f}$$

A linear integral equation for $\Theta_N(\tau)$ is obtained directly from (3.7*a, b*) and (3.8*f*),

$$b\Theta_N(\tau) = \int_0^\infty (1-\xi)\,\mathrm{e}^{-\xi}\,\Theta_N(\tau-\xi)\,\mathrm{d}\xi \quad \text{with} \quad b \equiv \frac{1}{\beta(\gamma-1)\,Q/C_p\,T_{N_o}}, \tag{3.8g}$$

with complex eigenvalues $\sigma = \sigma_r + \mathrm{i}\omega(\Theta_N \propto \mathrm{e}^{\sigma\tau})$ which are solutions of an algebraic equation,

$$\sigma^2 + (2-b^{-1})\,\sigma + 1 = 0, \tag{3.8h}$$

exhibiting a stability domain ($\sigma_r < 0$) characterized by $b^{-1} < 2$ with a Hopf bifurcation ($\sigma_r = 0$, $\omega = 1$) at $b^{-1} = 2$ representing the onset of galloping detonations.

By comparison, the progress variable of the square-wave model is an Heaviside function, $y = H(\xi - \xi_i(\tau))$ with a singular reaction rate obtained from (3.8*d*) in the form $w = -(\mathrm{d}\xi_i/\mathrm{d}\tau)\,\delta(\xi-\xi_i(\tau)) + \delta(\xi-\xi_i(\tau))$, where $\delta(\xi-\xi_i)$ is the Dirac function. Then by using results of the Appendix, $\mathrm{d}\xi_i/\mathrm{d}\tau = -\mathrm{d}\Theta_N(\tau-\xi)/\mathrm{d}\tau$, ($\delta m$ is neglected in (A 13)) one obtains from (3.7*a, b*) a difference–differential equation of the same advanced type as (3.6*b*) with $C = 0$,

$$b\Theta_N(\tau) = \mathrm{d}\Theta_N(\tau-1)/\mathrm{d}\tau. \tag{3.9a}$$

The comparison with (3.8*g*) is made easier by writing (3.9*a*) in the form

$$b\Theta_N(\tau) = \int_0^\infty \delta'(\xi-1)\,\Theta_N(\tau-\xi)\,\mathrm{d}\xi. \tag{3.9b}$$

Equations (3.9*a, b*) yield a transcendental equation for the eigenvalues

$$b = \sigma\,\mathrm{e}^{-\sigma}, \tag{3.9c}$$

presenting an infinite set of discrete unstable modes with unbounded amplification rates increasing with frequency ($\sigma_r \to +\infty$, $\omega \to \infty$). This is quite different from the spectrum obtained from (3.8*h*). Equations (3.8*g*) and (3.9*b*) both belong to the same class characterized by a distribution of reaction rate whose linear perturbation may be written in terms of the temperature fluctuation at the shock as

$$\delta\Omega(\xi,\tau) = G(\xi)\,\Theta_N(\tau-\xi) \Rightarrow b\Theta_N(\tau) = \int_0^\infty G(\xi)\,\Theta_N(\tau-\xi)\,\mathrm{d}\xi. \tag{3.10a}$$

The corresponding equation of eigenmodes is written in terms of the Laplace transform of $G(\xi)$ as

$$b = \int_0^\infty G(\xi)\,\mathrm{e}^{-\sigma\xi}\,\mathrm{d}\xi, \tag{3.10b}$$

where $b = O(1)$ is a coefficient given in (3.8*g*) and where, in general, $G(\xi)$ may be expressed in terms of the steady state solution, see (4.7*b*). The difference of results (3.8*h*) and (3.9*c*) illustrates the sensitivity of the detonation dynamics to the profile of the rate of heat release. Equation (3.10*b*) bridges the gap between the singular dynamics of detonation models with a discontinuous structure and the stability of detonations characterized by a sufficiently smooth inner structure. All the information is now collected to derive an asymptotic solution of the reactive Euler equations.

## 4. Asymptotic solution of the reactive Euler equations

### 4.1. *Basic approximation for gaseous detonations*

To begin we analyse the case

$$(\gamma-1) \ll 1. \tag{4.1a}$$

When the ratio of specific heats is sufficiently close to unity the isentropic modifications of temperature by the acoustic waves are less than heating by the chemical reaction (Blythe & Crighton 1989). When the pressure dependence of the reaction rate is neglected, entropy and species equations (2.3*c*, *d*) are no longer coupled to the equations of fluid mechanics except through the conditions at the shock,

$$\frac{\partial}{\partial \tau} T + m(\tau)\frac{\partial}{\partial \xi} T = \frac{Q}{C_p} w(T, y), \tag{4.1b}$$

$$\frac{\partial}{\partial \tau} y + m(\tau)\frac{\partial}{\partial \xi} y = w(T, y). \tag{4.1c}$$

These equations are easily solved by introducing the time lag $\Delta\tau(\xi, \tau)$ such that $\tau - \Delta\tau$ is the time at which a fluid particle which is located at the current time $\tau$ at position $\xi$, crossed the shock,

$$\xi = \int_{\tau-\Delta\tau(\xi,\tau)}^{\tau} m(\tau')\,\mathrm{d}\tau', \tag{4.1d}$$

$$\frac{\partial}{\partial \tau}\Delta\tau(\xi,\tau) + m(\tau)\frac{\partial}{\partial \xi}\Delta\tau(\xi,\tau) = 1. \tag{4.1e}$$

Using the boundary conditions at the shock ($\xi = 0$: $T = T_N$, $y = 0$) solutions of (4.1*b*, *c*) are written as

$$T - \frac{Q}{C_p} y = T_N(\tau - \Delta\tau(\xi, \tau)), \tag{4.1f}$$

$$\int_0^y \frac{\mathrm{d}y'}{\Phi(T_N(\tau-\Delta\tau(\xi,\tau)), y')} = \Delta\tau(\xi,\tau) \quad \text{where} \quad \Phi(T_N, y) \equiv w(T_N + yQ/C_p, y). \tag{4.1g}$$

The progress variable and the temperature given by (4.1*f*, *g*) are more conveniently written in terms of the steady state solution as

$$y = \mathscr{Y}_o(T_N(\tau-\Delta\tau(\xi,\tau)), \Delta\tau(\xi,\tau)), \quad T = \mathscr{T}_o(T_N(\tau-\Delta\tau(\xi,\tau)), \Delta\tau(\xi,\tau)) \tag{4.2a}$$

where $\mathscr{Y}_o(T_N, \xi)$ and $\mathscr{T}_o(T_N, \xi)$ are stationary solutions

$$\mathrm{d}\mathscr{Y}_o/\mathrm{d}\xi = w(\mathscr{Y}_o, \mathscr{T}_o), \quad \mathrm{d}\mathscr{T}_o/\mathrm{d}\xi = w(\mathscr{Y}_o, \mathscr{T}_o)\, Q/C_p \tag{4.2b}$$

satisfying the boundary condition

$$\xi = 0: \quad \mathscr{Y}_o = 0, \quad \mathscr{T}_o = T_N. \tag{4.2c}$$

Values of $y$, $T$ and $w$ at the current time $\tau$ and at position $\xi$, are thus expressed by the stationary distributions at position $\Delta\tau(\xi,\tau)$ with a shocked gas temperature $T_N$ at an earlier time $\tau-\Delta\tau(\xi,\tau)$. Once the distribution of the sound speed (2.5*d*) is expressed from (4.1*f*) and (4.2*a*) in terms of the steady distribution as

$$a/a_{N_o} = \mathscr{A}_o(T_N(\tau-\Delta\tau(\xi,\tau)),\Delta\tau(\xi,\tau)) \quad \text{where} \quad \mathscr{A}_o(T_N,\xi) \equiv (\mathscr{T}_0/T_{N_o})^{1/2}, \qquad (4.2d)$$

and when $p=\gamma^{-1}a^2\rho$ is introduced in (2.3*a*, *b*), equations for $\rho$ and $v$ are obtained in a closed form. Then, by eliminating $v$, the full problem of the detonation dynamics reduces to solve a nonlinear equation for $1/\rho$ with boundary conditions (2.6) and (2.7*a*, *b*). Further simplification is required for deriving an analytical solution.

### 4.2. *Distinguished limit*

From now on, let us define $\beta_N$ as the temperature sensitivity of the value of the reaction rate immediately downstream of the inert shock,

$$\beta_N \equiv \frac{1}{w_{N_o}} T_{N_o}\left[\frac{\partial}{\partial T} w\right]_{y=0,\,T=T_{N_o}}. \qquad (4.3a)$$

Guided by the order of magnitude estimates of §3, $b=O(1)$ in (3.8*g*), let us consider the following distinguished limits:

$$\beta_N \to \infty \quad \text{with} \quad \beta_N(\gamma-1)=O(1), \quad (\gamma-1)M_o^2=O(1), \quad Q/C_pT_{N_o}=O(1), \qquad (4.3b)$$

and focus the attention on small perturbations of the inert shock:

$$\frac{\delta M}{M_o}=O\left(\frac{1}{\beta_N}\right). \qquad (4.3c)$$

The condition $(\gamma-1)M_o^2=O(1)$ may be replaced without loss by $(\gamma-1)M_o^2 \gg 1$. Then, according to (2.6*a*–*e*), one has

$$\delta m = O\left(\frac{1}{\beta_N}\right), \quad \frac{\delta T_N}{T_{N_o}}=O\left(\frac{1}{\beta_N}\right), \quad \frac{\delta p_N}{p_{N_o}}=O\left(\frac{1}{\beta_N}\right), \quad \text{but} \quad \frac{\delta v_N}{v_{N_o}}=O(1). \qquad (4.3d)$$

At the leading order of (4.3*b*, *c*), the approximation $m=1$ is valid in (4.1*b*–*g*) yielding $\Delta\tau(\xi,\tau)=\xi$ and the distributions (4.2*a*–*c*) of $y$, $T$ and $w$ at the current time $\tau$ correspond simply to stationary distributions but with a gas temperature at the shock at an early time $\tau-\xi$,

$$y=\mathscr{Y}_o(T_N(\tau-\xi),\xi), \quad T-yQ/C_p=T_N(\tau-\xi). \qquad (4.3e)$$

Let us introduce a reduced fluctuation of the shocked gas temperature,

$$\Theta_N \equiv \beta_N(T_N-T_{N_o})/T_{N_o}=O(1), \qquad (4.3f)$$

and let $\Omega_o(\Theta_N,\xi)$ be the steady distribution of the rate of heat release solution of (4.2*b*, *c*). Perturbations (4.3*d*) are large enough to produce strong nonlinear effects in the limit (4.3*a*, *b*), $\delta w/w=O(1)$, yielding a leading order of the distribution of the unsteady solution written as

$$w=\lim_{\beta_N\to\infty}\Omega_o(\Theta_N(\tau-\xi),\xi). \qquad (4.3g)$$

This limit is discussed in §4.3.

According to (2.6*c*), the first conditions of (4.3*b*), $\beta_N(\gamma-1)=O(1)$, $(\gamma-1)M_o^2=O(1)$, yield a very subsonic flow immediately behind the shock,

$$M_{N_o}^2=O(1/\beta_N). \qquad (4.4a)$$

Notice that for an Arrhenius law (2.1$f$), one would have $\beta_N = E/RT_{N_o}$ and condition (3.6$e$) is automatically fulfilled by (4.4$a$) when $E/RT_{N_o} \gg 1$.

Equations (2.6$c$) and (2.6$g$) yield

$$M_{CJ}^2 \approx \frac{4}{(\gamma+1)} \frac{Q}{C_p T_N} [(\gamma-1) M_o^2 + 2], \tag{4.4b}$$

in such a way that the two last conditions of (4.3$b$), $(\gamma-1)M_o^2 = O(1)$, $Q/C_p T_{N_o} = O(1)$, correspond to a high degree of overdrive,

$$1/f \equiv M_{CJ}^2/M_o^2 = O(1/\beta_N), \tag{4.4c}$$

a condition that still holds for $(\gamma-1)M_o^2 \gg 1$ and, thus, for $M_{CJ}^2 \gg 1$. In both cases, $(\gamma-1)M_o^2 = O(1)$ and $(\gamma-1)M_o^2 \gg 1$, low overdrives ($f$ approaching 1) correspond to $Q/C_p T_{N_o} = O(\beta_N)$ in the distinguished limit $\beta_N \to \infty$, $\beta_N(\gamma-1) = O(1)$. Thus, according to (3.6$d$) and (4.4$a$–$c$), the distinguished limit (4.3$b$) ensures that a zero-Mach-number approximation, $M_1^2 = O(1/\beta_N)$ is valid within both the induction zone and the reaction region ($M_1 \equiv v/a$ is the local Mach number).

### 4.3. *Nonlinear integral equation for overdriven detonations*

The pressure term of (2.4) being negligible, $\delta p/p = O(1/\beta_N)$ across the detonation structure, the quasi-isobaric mass conservation (3.7$a, b$) is valid at the leading order of an asymptotic expansion in the limit (4.3$b, c$). By using the unsteady distribution of the rate of heat release obtained in (4.3$g$) this yields a nonlinear integral equation for $\Theta_N(\tau)$

$$1 + b\Theta_N(\tau) = \int_0^\infty \Omega_{o\infty}(\Theta_N(\tau-\xi), \xi)\,d\xi, \tag{4.5a}$$

where $b$ is the same coefficient of order unity as in (3.8$g$) in which $\beta$ is replaced by $\beta_N$ and where $\Omega_{o\infty}$ is, according to (4.3$g$), equal to

$$\Omega_{o\infty}(\Theta_N, \xi) \equiv \lim_{\beta_N \to \infty} \Omega_o(\Theta_N, \xi) \quad \text{with} \quad \Theta_N = O(1). \tag{4.5b}$$

With a uniform sensitivity to temperature of an Arrhenius law (2.1$f$), the limit (4.3$b$) used in (4.5$b$) yields a singular distribution leading to spurious singularities of the square-wave model (see §3),

$$\Omega_{o\infty} = \xi_i^{-1}(\tau-\xi)\,\delta(\xi/\xi_i(\tau-\xi)-1) \quad \text{with} \quad \xi_i(\tau) = \exp(-\Theta_N(\tau)). \tag{4.5c}$$

Such a brute force limit of an Arrhenius law, so useful in flame theory (see Clavin 1994 for a review), cannot be the right strategy to capture the threshold and the dynamics of galloping detonations. As shown by the particular examples studied in §3.4, a non-zero thickness of the region of heat release (finite rate of the exothermal reaction) is required for describing the dynamics in the high frequency range. When the distinguished limit (4.3$b$) is modified by using (4.1$f$) with a small heat release approximation,

$$\beta_N q = O(1) \quad \text{with} \quad q \equiv Q/C_p T_{N_o}, \tag{4.5d}$$

one gets at the leading order

$$\exp(-E/RT)/\exp(-E/RT_{N_o}) \approx \exp(\beta_N q y)\exp(\Theta_N(\tau-\xi)). \tag{4.5e}$$

A smooth distribution $\Omega_{o\infty}(\xi, \tau)$ is obtained from (4.1) and (4.2) in this limit. The time-dependent distribution so obtained is the same as (4.6$f$) with $\phi(y) = (1-y)\exp(\beta_N q y)$. The end result is similar to (4.5$c$) but with a smooth function $\Omega_o(\xi/\xi_i)$ replacing the

Dirac distribution $\delta(\xi/\xi_i - 1)$. A one-step kinetic model (2.1*f*) and assumption (4.5*d*) are not accurate approximations of ordinary detonations. However the end result, as given by (4.6*f*), is a fair representation of the unsteady structure of real detonations in which large variations of a smoothly distributed heat release rate are induced by small variations of the shocked gas temperature (see figure 2). This model (4.6*f*) picks up the essential features and will be used for simplicity in the rest of this paper. For large heat release it may be justified as follows. Contrary to flames, the chemical processes involved in the induction period are essential in the structure and dynamics of detonations and must be considered separately from the heat release rate. In the complex chemical networks of combustion, the elementary reactions involved in the induction period are different from those governing the heat release. They are athermal and extremely sensitive to temperature. The activation energies of the exothermic reactions are smaller. Within the framework of a crude modelling of the combustion rate by a one step overall reaction, one is led to consider two activation energies: $\beta_N$ defined by (4.3*a*) and governing the temperature sensitivity of the athermal induction period and $\beta_T$ governing the temperature sensitivity of the rate of heat release with $\beta_T \ll \beta_N$. A general model of reaction rate yielding a smooth distribution in the limit (4.3*b*, *c*) is

$$W(y,T) \propto t_{col}^{-1}\, h(T_N)\, g(T)\, (1-y)^n \quad \text{with by definition} \quad g(T_N) = 1 \qquad (4.6a)$$

and where $T_N$ has to be taken at retarded time $\tau - \xi$, $T_N\ (\tau - \xi)$, with

$$\beta_N \equiv h^{-1} T_N\, \mathrm{d}h/\mathrm{d}T_N \gg \beta_T \equiv g^{-1} T\, \mathrm{d}g/\mathrm{d}T, \qquad (4.6b)$$

so that

$$\beta_N \to \infty \quad \text{with} \quad (4.3b\text{–}c) \quad \text{and} \quad \beta_T = O(1). \qquad (4.6c)$$

By using the same reference time as in (2.2*b*) and following (4.1), one gets from (4.6*b*, *c*),

with

$$\left.\begin{aligned} &\int_0^y \frac{\mathrm{d}y'}{\xi_i^{-1}(\tau-\xi)\,\phi(y')} = \frac{\xi}{\alpha} \\ &\phi(y) \equiv (1-y)^n g(T_{N_o} + yQ/C_p), \quad \phi(y=0) = 1, \end{aligned}\right\} \qquad (4.6d)$$

where the reduced induction length $\xi_i(\tau) \equiv h(T_{N_o})/h(T_N(\tau))$ is a function of the gas temperature immediately downstream of the shock. From now on let us assume that $h(T_N)$ is an Arrhenius law,

$$h(T_N) \propto \exp(-E_N/RT_N) \Rightarrow \beta_N = E/RT_{N_o}, \quad \xi_i(\tau) = \exp(-\Theta_N(\tau)). \qquad (4.6e)$$

According to (4.6*d*–*e*), the time dependent distribution of the rate of heat release in (4.5*a*) is

$$\Omega_{o\infty}(\xi,\tau) = \frac{1}{\xi_i(\tau-\xi)}\,\Omega_o\!\left(\frac{\xi}{\xi_i(\tau-\xi)}\right) \quad \text{with} \quad \int_0^\infty \Omega_o(\zeta)\,\mathrm{d}\zeta = 1, \qquad (4.6f)$$

where $w = \Omega_o(\xi)$ is the reduced distribution of the steady state,

$$\Omega_o = \frac{\mathrm{d}y_o}{\mathrm{d}\xi} \quad \text{with} \quad \int_0^{y_o} \frac{\mathrm{d}y}{\phi(y)} = \frac{\xi}{\alpha}, \qquad (4.6g)$$

and where the choice of $\alpha$ determines the unity length. The most popular scaling is such that $y = \frac{1}{2}$, for $\xi = 1$:

$$\int_0^{1/2} \frac{\mathrm{d}y}{\phi(y)} = \frac{1}{\alpha}. \qquad (4.6h)$$

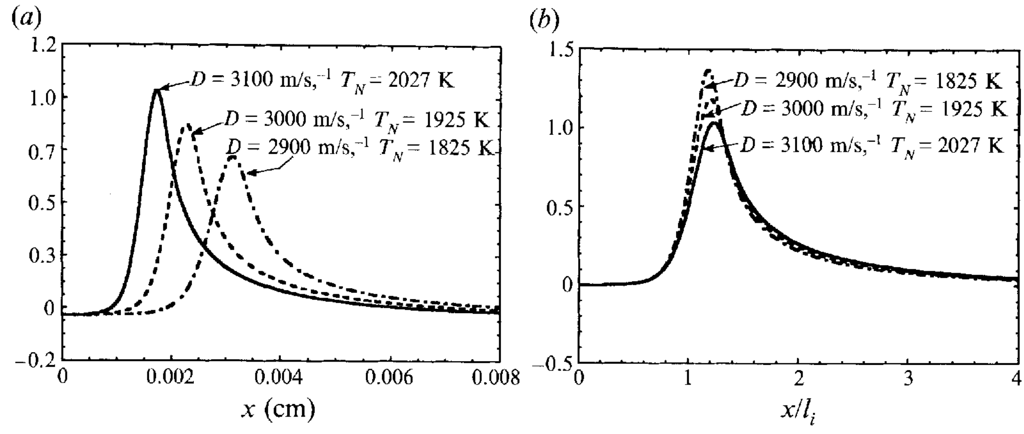


FIGURE 2. Distributions of heat release rate computed with a complex chemical network for plane detonations propagating at constant speed in hydrogen–oxygen mixtures at a stoichiometric composition and ordinary conditions, $T_u = 298$ K, $p_u = 1$ atm, for three different detonation speeds. (*a*) Reduced heat release rates $\Omega_o(x)$ as a function of the distance from the shock and arbitrarily normalized by the maximum value of the case corresponding to $D = 3100$ m s$^{-1}$. (*b*) The same results plotted in the form $l_i^{-1}\Omega_o(x/l_i)$, where length $l_i$ is defined here as the position of the maximum value in (*a*). The assumption of a self-similar deformation law (4.6*f*) is found to be verified in this real mixture with a uniform accuracy better than 30% for $\delta D/D \approx 10\%$.

A numerical computation of the detonation structure with a detailed chemical kinetics scheme for a $H_2$–$O_2$ mixture shows that (4.6*f*) is a fair approximation (see figure 2). Then, according to (4.5*a*) and (4.6), the evolution equation describing the nonlinear dynamics of gaseous overdriven detonations is

$$1+b\Theta_N(\tau) = \int_0^\infty \exp(\Theta_N(\tau-\xi))\,\Omega_o(\xi\exp(\Theta_N(\tau-\xi)))\,\mathrm{d}\xi$$

with

$$b^{-1} \equiv \beta_N(\gamma-1)\frac{Q}{C_p T_{N_o}} = O(1) \qquad (4.7a)$$

yielding, in the linear approximation, the same equation as (3.10) with

$$G(\xi) = \Omega_o(\xi)+\xi\,\mathrm{d}\Omega_o(\xi)/\mathrm{d}\xi = \mathrm{d}(\xi\Omega_o)/\mathrm{d}\xi. \qquad (4.7b)$$

Equations (4.7*a, b*) exhibit the influences of both the distribution of heat release $\Omega_o(\xi)$ and the sensitivity of the induction kinetics to temperature $\beta_N$ through the coefficient $b$.

### 4.4. *Generalized Arrhenius model*

Most of the existing numerical results have been obtained with an Arrhenius law (2.1*f*). For the sake of comparison with the existing results, it is convenient to carry out a parametric investigation of solutions of (4.7*a*) with a kinetic law satisfying assumptions (4.6*a, b*) and yielding for $\beta_N = \beta_T$ the same steady distribution $\Omega_o(\xi)$ as that obtained by an Arrhenius law. Consider the kinetic model given by

$$W(y,T) = B\exp\left(-\frac{E_N}{RT_N}\right)\exp\left\{-\beta_T\left(\frac{T_N}{T}-1\right)\right\}(1-y), \qquad (4.8a)$$

where $\beta_T$ is a parameter different from $\beta_N$. This law satisfies (4.6) and reduces to (2.1*f*) when $\beta_T = \beta_N \equiv E_N/RT_N$. Following the analysis of the preceding section with $T_N\ (\tau-\xi)$ introduced in (4.8*a*), one gets an evolution equation (4.7*a*) in which the steady distribution $\Omega_o(\xi)$ is obtained from (4.6*g*) with

$$\phi(y) = (1-y)\exp\left(-\beta_T q \frac{y}{1+qy}\right), \quad q \equiv \frac{Q}{C_p T_{N_o}}. \tag{4.8b}$$

The parameter of order unity $q$ characterizes the temperature increase due to the heat release. This parameter appears both in $\Omega_o(\xi)$ through $\phi(y)$ and in the dimensionless parameter $b^{-1} \equiv \beta_N(\gamma-1)\, q$ of (4.7*a*). The larger is the quantity $\beta_T q = O(1)$, the stiffer is the reduced distribution of the rate of heat release $\Omega_o(\xi)$. To summarize, two essential parameters control the final results of (4.7*a*) and (4.8*a*), $b^{-1} \equiv \beta_N(\gamma-1)\, q$ and $\beta_T q$; the parameter $q$ left in (4.8*b*) has a minor influence. Three points have to be kept in mind:

(i) The same steady distribution $\Omega_o(\xi)$ as that given by an Arrhenius law is obtained when $\beta_T = \beta_N \equiv E_N/RT_{N_o}$;

(ii) the exponential model of §3.4 corresponds to $\beta_T = 0$,

(iii) the limit $\beta_T \rightarrow \infty$ yields the square-wave model.

## 5. Discussion of the results

### 5.1. *Linear spectrum*

The spectrum of the linearized equation (4.7*a*) corresponds to the roots of (3.10*b*) with (4.6*g*), (4.7*b*) and (4.8*b*). Typical unstable spectra are plotted in figure 3 for different sets of parameters ($b^{-1}$, $\beta_T$, $q$). These unstable spectra are qualitatively similar to those obtained numerically by a shooting method as in Lee & Stewart (1990) from equations (2.3) with an Arrhenius law (2.1*g*). For the sake of comparison, such spectra are also plotted in figure 3 for conditions corresponding to the same sets of parameters ($b^{-1}$, $\beta$, $q$) but with more realistic values of the overdrive factor than those satisfying the conditions of validity of the quasi-isobaric approximation. These spectra exhibit a finite number of unstable modes and the high-frequency modes are stable. Both the growth rate and the frequency of the most unstable mode increase with $\beta_T$ and the unbounded spectrum of the square-wave model is recovered in the limiting case $\beta_T \rightarrow \infty$. Quantitative differences in growth rates and frequencies of order $M_{N_o} \approx (\gamma-1)^{1/2}$ are observed between (4.7*a*, *b*) and numerical analysis of linearized equations (2.3). These differences result from the effects of acoustic waves which have been neglected here; we postpone this discussion to the last section.

The stability limits are plotted in figure 4 in a ($b^{-1}$, $\beta_T$) plane. The results obtained from (4.7*a*, *b*) with (4.8*a*, *b*) for $q = 1.2$ are compared with those obtained from the numerical study of the spectrum of the linearized equations (2.3) with (5.1*a*) for $\gamma = 1.2$ and $q = 1.2$. Both stability limits correspond to a super critical Hopf bifurcation. The two marginal stability curves are close and correspond roughly to a straight line crossing the points ($b^{-1} = 2$, $\beta_T = 0$) and ($b^{-1} = 0$, $\beta_T = 12$). Increasing one of the parameters $b^{-1}$ or $\beta_T$ has a tendency to destabilize the detonation. Here also a discrepancy of the same order of magnitude as $M_{N_o} \approx (\gamma-1)^{1/2}$ is observed in the frequency of the marginal mode.

Let us now investigate the phase space by increasing $b^{-1}$ for different fixed values of $\beta_T$:

(i) Consider first the domain $\beta_T < 6$. Here the first bifurcation concerns the lowest frequency mode of the spectrum and occurs at a critical value of $b^{-1}$ ranging from 1 to 2 when $\beta_T$ decreases from 6 to 0. In the unstable domain, the growth rate of this

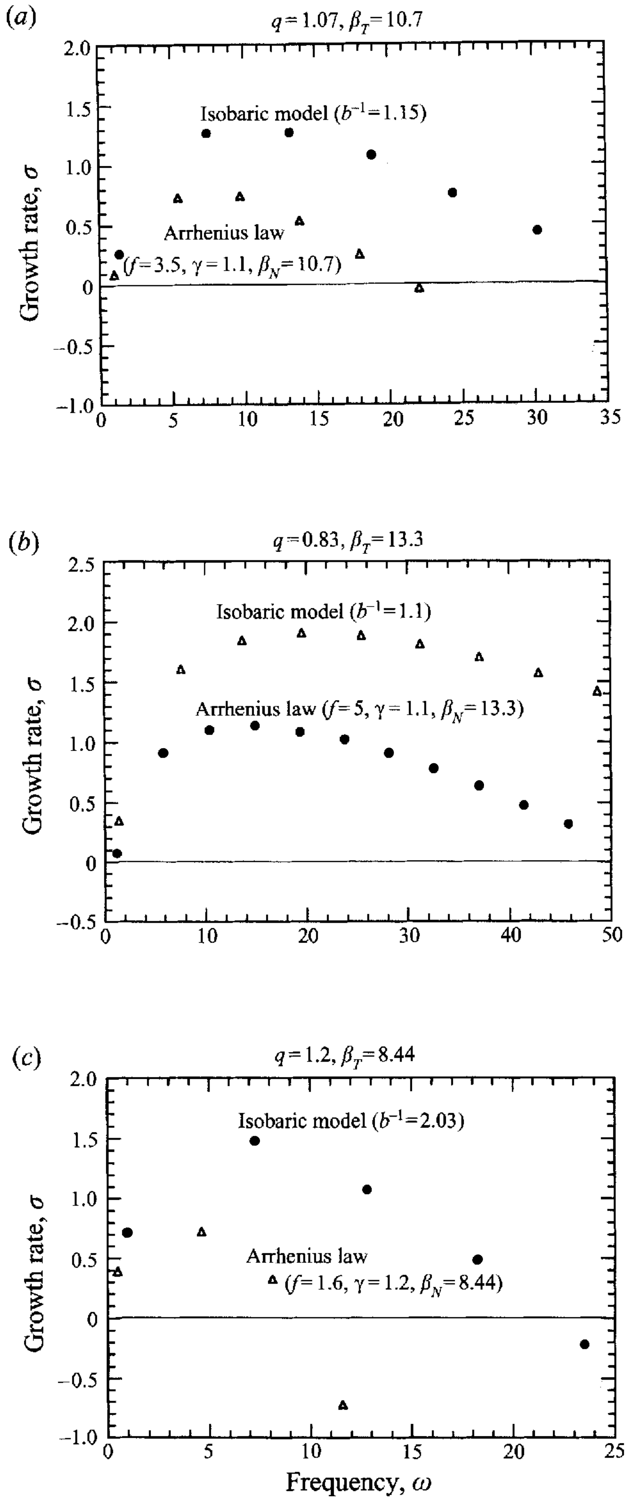


FIGURE 3. Growth rate *vs.* frequency for eigenvalues of typical unstable spectra of detonations corresponding to model (4.7*a*) with (4.8*a*, *b*) and full equations (2.3) with (2.1*f*), respectively. The timescale is here the half reaction time $t_{1/2}$.

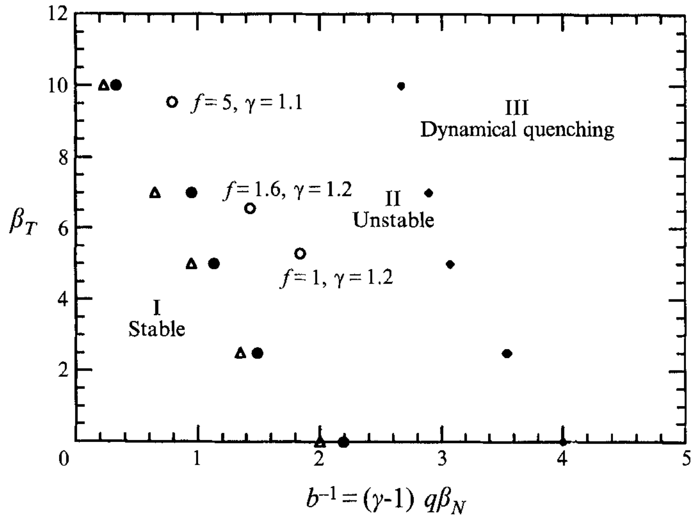


FIGURE 4. Stability limits in a plane $(b^{-1}, \beta_T)$. The detonation is stable in region I, unstable in region II and a dynamical quenching is predicted in region III. △, stability limits obtained by solving (4.7 *a, b*) with a reaction model (4.8 *a, b*); ●, stability limits obtained by solving the exact linear problem with the same reaction model; ○, stability limits obtained by solving the exact linear instability problem with an Arrhenius law and different values of the degree of overdrive, the same limits are obtained by DNS; ◇, limits at which the frequency of the first mode (fundamental) becomes zero.

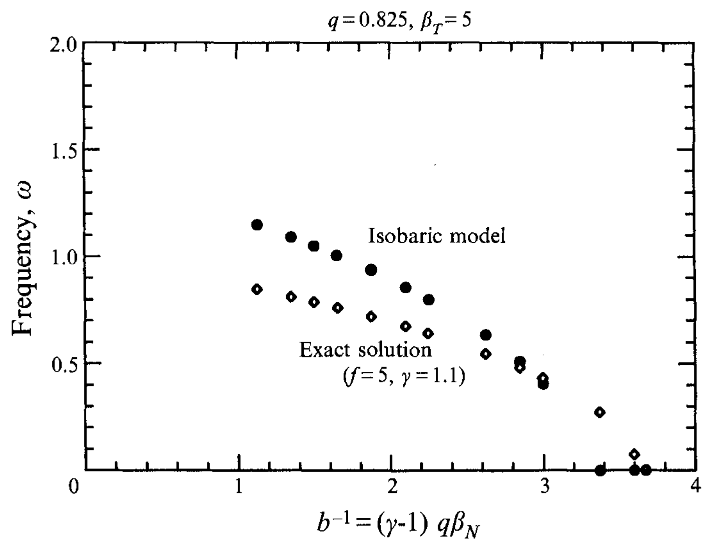


FIGURE 5. Variation of the frequency as a function of parameter $b^{-1} \equiv \beta_N(\gamma-1)\,q$. The timescale is the half reaction time $t_{1/2}$.

linear mode increases with $b^{-1}$ while its frequency decreases toward zero, see figure 5. This frequency reaches zero at another critical value of $b^{-1}$ which varies from 2.8 to 4 when $\beta_T$ decreases from 6 to 0 (see figure 4). This second critical limit is also plotted in figure 4. In the particular case $\beta_T = 0$ there is, according to (3.8*h*), only one oscillatory mode in the unstable spectrum for $2 < b^{-1} < 4$; the Hopf bifurcation occurs at $b^{-1} = 2$, and the frequency of the unstable mode reaches zero in the unstable domain at $b^{-1} = 4$. A similar scenario occurs in all the domain $0 < \beta_T < 6$; no other linear

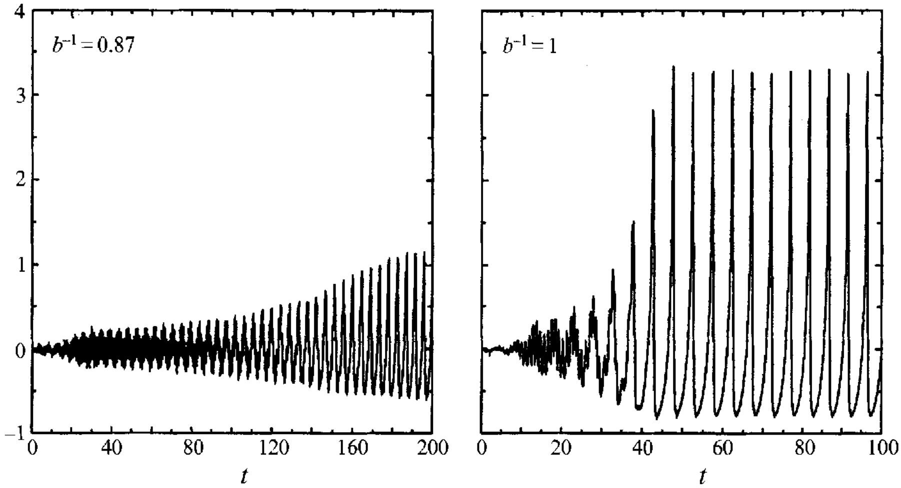


FIGURE 6. Evolution of $\Theta_N(t)$ obtained from (4.7*a*) and (4.8*a*) for $\beta_T = 7$ and two values of $b^{-1} = 0.87$ and 1. The timescale is the half reaction time $t_{1/2}$. For these parameters, the linear stability equation (4.7*b*) yields two unstable modes: $\sigma + \mathrm{i}\omega = 0.035 + \mathrm{i}\ 1.41$ and $0.142 + \mathrm{i}\ 6.85$ for $b^{-1} = 0.87$, $\sigma + \mathrm{i}\omega = 0.12 + \mathrm{i}\ 1.32$ and $0.25 + \mathrm{i}\ 6.86$ for $b^{-1} = 1$. Notice that the most unstable linear mode develops first but the final periodic oscillation has a period equal to that of the fundamental mode.

mode becomes unstable in the unstable domain of the phase space between the two above mentioned critical values of $b^{-1}$.

(ii) For $\beta_T > 6$, the first bifurcation which appears by increasing $b^{-1}$ concerns a higher frequency mode; the linear mode with the lowest frequency bifurcates at a higher value of $b^{-1}$. Thus, as shown in figure 3, the spectrum presents several unstable modes. The frequency of the fundamental mode (lowest frequency) still decreases when increasing further $b^{-1}$ and reaches zero on a critical line plotted in figure 4; for example this second critical value of $b^{-1}$ is about 2.7 for $\beta_T = 10$.

### 5.2. *Nonlinear dynamics*

Numerical solutions of (4.7*a*) may be obtained as the response to external perturbations by using a direct extension of (4.7*a*) to the case where perturbations are present in the fresh mixture in which the detonation propagates,

$$1 + b[\Theta_N(\tau) - \Theta_{N_o}(\tau)] = \int_0^\infty \exp(\Theta_N(\tau - \xi))\, \Omega_o(\xi \exp(\Theta_N(\tau - \xi)))\, \mathrm{d}\xi, \qquad (5.1)$$

where $\Theta_{N_o}(\tau)$ are the induced fluctuations of the shocked gas temperature immediately downstream of the inert shock propagating with a velocity $D_o$ across the external perturbations, as given by Rankine–Hugoniot conditions (2.6). The calculation is initialized at $\tau = 0$ by using the unperturbed steady state solution for $\tau < 0$, $\Theta_N(\tau) = \Theta_{N_o}(\tau) = 0$, and a prescribed perturbation $\Theta_{N_o}(\tau) \neq 0$ for $0 < \tau < \Delta$. For sufficiently small perturbations the asymptotic time behaviour of the solution of (5.1) does not depend on $\Theta_{N_o}(\tau)$ (at least in a statistical sense when the solution is chaotic); the so-obtained function $\Theta_N(\tau)$ may be considered as an intrinsic solution of (4.7*a*). In the unstable region of the phase space, between the two critical values of $b^{-1}$ mentioned above, oscillatory solutions are observed. The period of these two critical values of $b^{-1}$ is either that of the unstable mode with the lowest frequency or of its subharmonics (see figures 6 and 7). When $b^{-1}$ is increasing into the unstable domain from the marginal

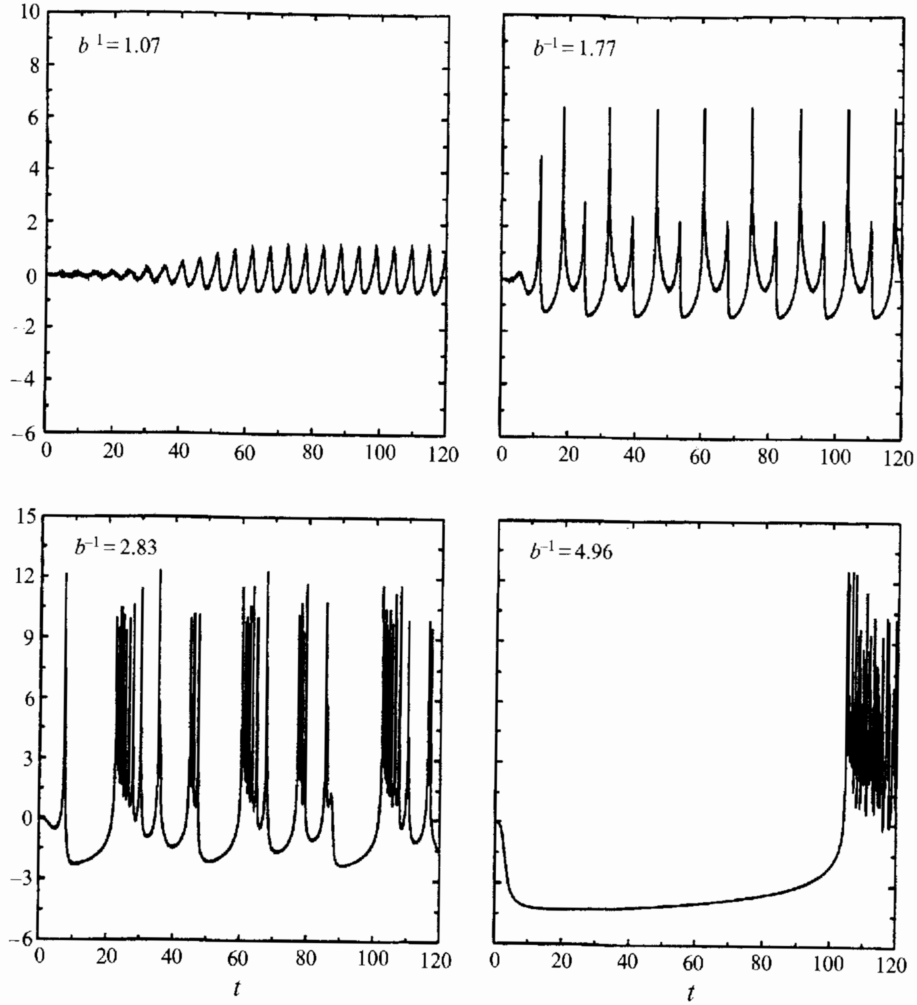


FIGURE 7. Evolution of $\Theta_N(t)$ obtained from (4.7*a*) and (4.8*a*) for $\beta_T = 5$ and four values of $b^{-1} = 1.07$, 1.77, 2.83 and 4.96. The timescale is the half reaction time $t_{1/2}$.

stability limit, the amplitude of the oscillatory solutions of (4.7*a*) increases. The solution experiences period doubling and a transition to chaos appears in the neighbourhood of the second critical value of $b^{-1}$ at which the frequency of the fundamental mode reaches zero (see figures 6 and 7). This chaotic behaviour is intermittent presenting bursts of high-frequency oscillations with large fluctuations of fast velocities ($\Theta_N > 0$) separated by wide periods of time in which the detonation propagates at slow motion with a quasi-constant speed much below the steady state one ($\Theta_N < 0$). The life-time of such a slow propagation regime increases very quickly with $b^{-1}$ while the quasi-constant propagation speed and the gas temperature $\Theta_N$ decrease (see figure 7). Such a scenario is similar for every value of $\beta_T$ at least in the domain which has been investigated, $0 < \beta_T < 12$. The presence of additional unstable modes at higher frequencies (when $\beta_T > 6$) influences neither the frequency of the nonlinear oscillations which is systematically governed by the fundamental mode, nor the scenario of transition to chaos. In particular the transition to chaos occurs as a rule in the neighbourhood of the points of the phase space at which the frequency of the fundamental mode reaches zero.

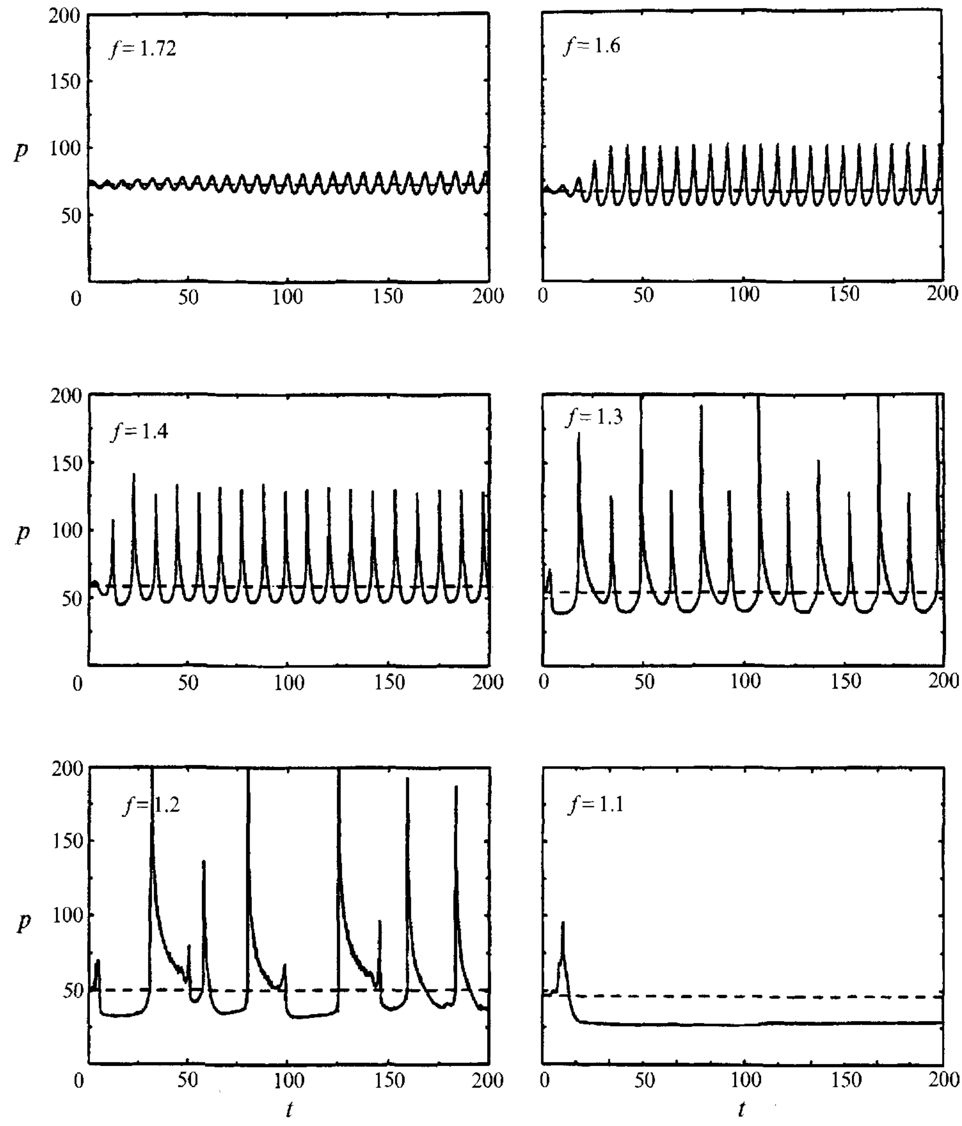


FIGURE 8. Evolution of the pressure at the Neumann spike (normalized by the initial pressure $p_u$) obtained by direct numerical simulations for piston-supported detonations with different values of the degree of overdrive and a fixed value of the activation energy in the Arrhenius law. The timescale is the half reaction time $t_{1/2}$.

Comparisons with direct numerical simulations (DNS) of planar detonations sustained by an Arrhenius law have also been carried out by using the same numerical method as in He & Clavin (1994). Good agreements are obtained at high overdrives and for a ratio of specific heats close to unity. The similarity of the nonlinear scenarios experienced by the solutions of (4.7*a*) and of direct numerical simulations (DNS) is striking even at low overdrives (see figure 8), but the amplitudes of highly nonlinear regimes in DNS (and more particularly of pressure fluctuations) at low overdrives are much larger than the ones considered by the limit (4.3*b*–*d*). For the sake of comparison, the transitions between the different regimes shown by DNS may be plotted in a $(b^{-1}, \beta_T)$-plane with $\beta_T = \beta_N$. In the case of a piston-driven detonation with a prescribed Arrhenius law, a decrease of the degree of overdrive corresponds to an increase of parameter $b^{-1}, f^{-1} \approx 2(\gamma-1)q$ and $b^{-1} \equiv \beta_N(\gamma-1)q$. The stability limits are shown in figure 4. The agreement with the results obtained from (4.7*a*) is satisfactory, however quantitative discrepancies appear at low overdrives.

Long periods of slow propagation are observed in figure 8 as they were in figure 7. They have been interpreted by He & Lee (1995) as responsible of dynamical quenching. The explanation is as follows: the complex chemical networks governing the combustion kinetics present cross-over temperatures defining the flammability limit as in $H_2$–$O_2$ mixtures (see for example He & Clavin 1993); if, during a period of deceleration, the shocked gas temperature immediately downstream of the inert shock drops below this cross-over temperature the induction time increases so much that the least external perturbation will quench the detonation. The critical conditions for such a dynamical quenching correspond roughly to the line of zero frequency of the fundamental mode in figure 4.

## 6. Conclusions and perspectives

We have derived a nonlinear integral equation for the dynamics of overdriven detonations as an asymptotic solution of the reactive Euler equations in the distinguished limit (4.3*b, c*) with a kinetic model yielding (4.6*f, g*). Two approximations in (4.3*b, c*) are accurate for ordinary gaseous mixtures: a high-temperature sensitivity of the induction period, $\beta_N \gg 1$, and a ratio of specific heats close to unity, $(\gamma-1) \ll 1$. Consequently the reversible pressure fluctuations have negligible effects on the entropy waves (2.3*c*). Thus, the main restriction of the results obtained is the last condition of (4.3*b, c*) limiting the analysis to piston-driven gaseous detonations at high overdrives. Such a limit yields a quasi-isobaric approximation which is useful to shed light on the basic mechanisms of galloping detonations even outside the domain of validity of the analysis. The approximation of a small local Mach number is generally accurate immediately downstream of the shock, typical values are $M_{N_o} \approx 0.2$–$0.3$ for CJ waves. However, the local Mach number increases with the heat release to reach unity in the burned gases of the marginal CJ detonations ($f = 1$). Pressure effects can no longer be neglected in the detonation dynamics at low overdrives. They introduce quantitative but no qualitative differences in a wide range of ordinary regimes of propagation. An analysis of these effects are presented elsewhere (Clavin & He 1995; He 1995). The main features are as follows: fluctuations of the heat release being governed by the shocked gas temperature with a delay introduced by the propagation of the entropy waves, the characteristic time of evolution is longer than the transit time of the acoustic wave propagating downstream by a factor $M_{N_o}^{-1}$. Owing to the small value of $M_{N_o}$, a strong amplification of acoustic waves by a coherent mechanism as in thermo-acoustic instabilities cannot occur. The role of the acoustic wave propagating upstream across the detonation structure is limited to bring the information back to the shock. This increases the delay in (4.7*a*) yielding a correction of the same order of magnitude as $M_{N_o}$ without modifying the form of the equation. The marginal CJ case remains an open problem.

## Appendix

The reduced induction length $\xi_i(\tau)$ corresponds to (2.2) with $x = x_r(t)$,

$$\xi_i(\tau) = \frac{1}{t_{ref}\rho_u D_0}\int_{x_s(t)}^{x_r(t)} \rho\,\mathrm{d}x, \quad \tau = \frac{t}{t_{ref}}. \tag{A 1}$$

By choosing the reference time equal to the unperturbed induction time, $t_{ref} = t_{i_o}$, the exothermic sheet of the unperturbed state is located at $\xi = 1$,

$$t_{ref} = t_{i_o} \equiv \frac{l_{i_o}}{v_{N_o}}, \quad \xi_{i_o} = 1, \quad \delta\xi_i = \frac{\delta l_i}{l_{i_o}} + \int_0^1 \frac{\delta\rho}{\rho_{N_o}}\mathrm{d}\xi. \tag{A 2}$$

According to (2.5a–d) and (2.3*c*), the acoustic waves and the entropy waves propagating across the uniform induction zone ($0 < \xi < \xi_i$: $w = 0$) yield

$$\left.\begin{aligned} \delta p + \rho_{N_o} a_{N_o}\delta v = \delta f_+\left(\tau - \frac{\xi}{M_{N_o}^{-1}+1}\right), \quad \delta p - \rho_{N_o} a_{N_o}\delta v = \delta f_-\left(\tau + \frac{\xi}{M_{N_o}^{-1}-1}\right), \\ \delta p - a_{N_o}^2\delta\rho = \delta h(\tau - \xi), \end{aligned}\right\} \tag{A 3}$$

with $M_{N_o} < 1$. The linearized Rankine–Hugoniot conditions (2.6) at $\xi = 0$, imply that all the three functions $\delta f_+(\tau)$, $\delta f_-(\tau)$, $\delta h(\tau)$, are proportional to the perturbation of the reduced mass flux across the shock $\delta m(\tau)$, ($m = 1 + \delta m(\tau)$). Then, the perturbations $\delta p_r(\tau)$, $\delta v_r(\tau)$, $\delta\rho_r(\tau)$ at the entrance of the reaction sheet and at the current time $\tau$ are obtained from (A 3) at $\xi = \xi_{i_o} = 1$, as a linear combination of the three quantities of (3.1). By definition and by analogy with (2.3*f*) the reduced mass flux crossing the quasi-steady reaction sheet appearing in (3.3) is

$$m_r(\tau) \equiv \frac{\rho_r(\tau)}{\rho_u D_0}[u_r(\tau) + D_r(\tau)] \tag{A 4}$$

where $D_r(t) \equiv -\mathrm{d}x_r/\mathrm{d}t$ is the velocity of the reacting layer (see figure 1), $D_r(t) = D_o + \delta D_r(t)$, $m_r(\tau) = 1 + \delta m_r(\tau)$. Kinematic considerations and definitions (A 1) (A 4) and (2.3*f*) yield

$$\mathrm{d}l_i/\mathrm{d}t = \delta D(t) - \delta D_r(t), \quad \mathrm{d}\xi_i/\mathrm{d}\tau = \delta m(\tau) - \delta m_r(\tau). \tag{A 5}$$

On the other hand, owing to kinetic laws (2.1*f*), $\mathrm{d}l_i/\mathrm{d}t$ is governed by the fluctuations of the thermodynamic variables inside the induction zone. Solution of this problem from the basic equations is not an easy task in general, it would require at least the solution of the nonlinear Clarke equation (see §9.5 in Clarke 1985). An asymptotic solution may be found in the limiting case $(\gamma - 1) \ll 1$ for which equations (2.3*c, d*) reduce to (4.1b, c). The square-wave model is obtained from an Arrhenius law (2.1*f*) in the limit $\beta \equiv E/RT_{N_o} \to \infty$ and the induction length is determined by the runaway of the first term of the asymptotic expansion of the temperature,

$$\frac{T - T_N}{T_{N_o}} = \frac{\Theta}{\beta} + O\left(\frac{1}{\beta^2}\right), \quad y = O\left(\frac{1}{\beta}\right), \tag{A 6}$$

solution of the following equation:

$$\frac{\partial}{\partial\tau}\Theta + m(\tau)\frac{\partial}{\partial\xi}\Theta = t_{ref}\beta\frac{Q}{C_p T_{N_o}}B\exp\{-E/RT_N(\tau)\}\exp\Theta. \tag{A 7}$$

Let us choose for convenience the reference time as the induction time of the unperturbed solution:

$$t_{ref}^{-1} = \beta \frac{Q}{C_p T_{N_o}} B \exp(-E/RT_{N_o}). \tag{A 8}$$

Then, according to (4.1*d*) and (4.1*g*), the induction length, $\xi_i(\tau)$, and the time delay, $\Delta\tau(\xi_i, \tau)$, are solutions of the two coupled equations:

$$\xi_i(\tau) = \int_{\tau - \Delta\tau(\xi_i, \tau)}^{\tau} m(\tau')\,\mathrm{d}\tau', \tag{A 9}$$

$$\int_0^\infty \mathrm{d}\Theta' \exp(-\Theta') = 1 = \Delta\tau(\xi_i, \tau) \exp\left(\frac{E}{RT_{N_o}}\right) \exp\left(-\frac{E}{RT_N(\tau - \Delta\tau(\xi_i, \tau))}\right) \tag{A 10}$$

yielding for the unperturbed solution characterized by $m_o = 1$ and $T_N(\tau) = T_{N_o}: \Delta\tau_o = \xi_{i_o} = 1$. In the linear approximation, the time derivative of (A 9) yields

$$\frac{\mathrm{d}}{\mathrm{d}\tau}\xi_i(\tau) = \delta m(\tau) - \delta m(\tau - 1) + \frac{\mathrm{d}}{\mathrm{d}\tau}\Delta\tau(\xi_i(\tau), \tau) \tag{A 11}$$

where $\mathrm{d}\Delta\tau/\mathrm{d}\tau$ is obtained from the time derivative of (A 10),

$$\beta \frac{1}{T_{N_o}} \frac{\mathrm{d}T_N(\tau - 1)}{\mathrm{d}\tau} + \frac{\mathrm{d}}{\mathrm{d}\tau}\Delta\tau(\xi_i(\tau), \tau) = 0. \tag{A 12}$$

Equations (A 11) and (A 12) lead to

$$\frac{\mathrm{d}}{\mathrm{d}\tau}\xi_i(\tau) = \delta m(\tau) - \delta m(\tau - 1) - \beta \frac{1}{T_{N_o}} \frac{\mathrm{d}}{\mathrm{d}\tau} T_N(\tau - 1) \tag{A 13}$$

to give in dimensional form

$$\frac{1}{l_{i_o}} \frac{\mathrm{d}}{\mathrm{d}t} l_i(t) = \frac{\delta D(t) - \delta D(t - t_{i_o})}{l_{i_o}} - \beta \frac{1}{T_{N_o}} \frac{\mathrm{d}}{\mathrm{d}t} T_N(t - t_{i_o}) \quad \text{with} \quad \beta \gg 1. \tag{A 14}$$

Equation (3.4) is obtained from (A 5) and (A 13). When $\delta m$ is neglected in (A 13), one gets (4.5*c*).

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