

# Spinning Instability of Gaseous Detonations

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## Abstract

We investigate hydrodynamic instability of a steady planar detonation wave propagating in a circular tube to three-dimensional linear perturbations, using the normal mode approach. Spinning instability is identified and its relevance to the well-known spin detonation is discussed. The neutral stability curves in the plane of heat release and activation energy exhibit bifurcations from a low-frequency to high-frequency spinning modes as the heat release is increased at fixed activation energy. With a simple Arrhenius model for the heat release rate remarkable qualitative agreement is found with experiments that vary the mixture dilution, initial pressure, and tube diameter. The analysis contributes to an explanation of the spin detonation which has essentially been absent since the discovery of the phenomenon over seventy years ago.

## 1 Introduction

Spin detonation in gases is one of the forms of multi-dimensional detonation propagation. Its discovery was made by Campbell and Woodhead (1926, 1927) and dates back as early as 1926. They observed that detonation in tubes of circular cross section in a stoichiometric mixture of carbon monoxide and oxygen exhibits a highly luminous region which traces a helical path along the periphery of the tube at a nearly constant angular frequency. During its long history the spin phenomenon has been investigated in much detail experimentally using photographic and smoke-foil techniques (see e.g. Voitsekhovskii, Mitrofanov & Topchian 1963; Schott 1965). The structure of the spinning wave close to the wall has been well established and is shown to represent complex Mach configurations consisting of shock and detonation fronts and tangential discontinuities. Only a few studies have been devoted to the study of the spin internal structure. Rigorous theoretical and numerical studies of the spin are lacking, since the spin detonation is inherently three-dimensional and thus challenging. Spinning waves can be observed in slow combustion and their theoretical analyses exist in the literature, see, for example, Sivashinsky (1981), Matkowsky & Olagunju (1982). Also, spin detonation is frequently observed in two-phase mixtures, see, for example, Zhang & Gronig (1991). In this paper we focus on spin detonation in gases.

The spin is an important regime of multi-dimensional detonation, both from theoretical and practical points of view. From a theoretical perspective it can be considered as one of

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the manifestations of a cellular detonation. From practical considerations, the importance of knowing the mechanism of spinning detonation stems from the fact that as a mainly marginal phenomenon it is accompanied by localized regions of extremely high pressure (e.g. about  $170 p_0$  behind the spin head is reported in Voitsekhovskii et al. (1963), where  $p_0$  is the pressure in the fresh mixture). Obviously, such high pressures are to be avoided in many practical systems. In addition, practical detonation waves often occur subject to confinement and instability predictions obtained for detonation in infinite space may have limited applicability to such conditions. In particular, the spinning instability may have relevance to the problem of the pulse-detonation engine which uses detonations that propagate in cylindrical tubes. Therefore it is important to determine the effect of confinement on the instability boundaries. The strategy developed below is for a cylindrical geometry, but following similar ideas rectangular geometry can be handled without difficulty.

Despite the apparent significance of the problem, current understanding of the spinning detonation is at the level of qualitative speculations, there exist a clear need for a detailed look at fundamental mechanisms such as the instability of perturbations to initially steady, planar detonation. It is now well known that for a wide range of parameters one-dimensional detonations are unstable to both one- and two-dimensional linear perturbations (see e.g. Erpenbeck 1964, Lee & Stewart 1990, Bourlioux & Majda 1992, Short & Stewart 1998). This kind of instability has an important relationship to a cellular structure of multi-dimensional detonations. Except for Pukhnachev's early work (1963), all the existing literature on detonation stability that we are aware of is concerned with detonations extending to infinity in both the longitudinal and lateral directions. Pukhnachev (1963) sets down the modal problem for a detonation in a tube, but his work is incomplete in both the formulation and results. Importantly, no mention of spin detonation is made.

The main phenomenological characteristics of the spin detonation are found in the experimental literature. The structure of spin detonation was most carefully studied by Voitsekhovski et al. (1963) and independently by Schott (1965). Using photographic and smoke-foil techniques they investigated the complex details of the structure of the spinning front near the wall. The spin structure as proposed by Voitsekhovski et al. (1963) and Schott (1965) is schematically shown on figure 1(b). The dark bands on Fig. 1(a), corresponding to the spin head DE on figure 1(b), show schematically the smoke foil records left by a single-head spin detonation. In the case of multiple-head spin, several transverse waves similar to ABCDE exist, which can rotate in opposite direction and interact with each other creating cellular imprints on the soot-foils.

The experiments by Bone, Fraser & Wheeler (1935), and Gordon, Mooradian & Harper (1959) have shown that spin is a near-limit phenomenon occurring mostly in mixtures near their detonability limits. Since the spinning wave moves along a helical path, one of the easily measured characteristics of the spin is the pitch  $p$  of the helix, which is the axial advancement of the detonation shock complex for one rotation of the spin head. It was shown that for a given mixture composition the ratio of the pitch to the tube diameter  $d$ , i.e.  $p/d$ , is independent of the diameter and is about three for most mixtures. When the detonation is initiated by a strong source which leads to initial overdrive, it was observed that the occurrence of a spinning wave coincided with the detonation velocity approaching the Chapman-Jouguet (CJ) value. Thus an overdrive is likely to suppress spinning instabilities, similar to its well-known effect on longitudinal as well as transverse instabilities. Gordon

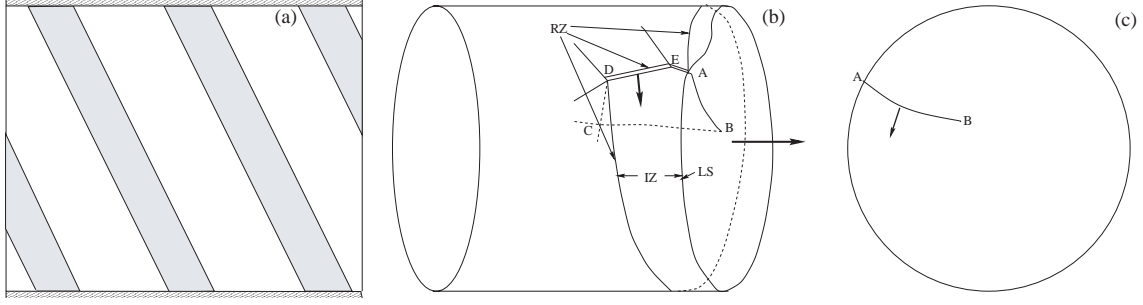


Figure 1: (a) Sketch of a soot foil record left by a single-head spin detonation; (b) schematic structure of the spin as proposed in Voitsekhovski et al. (1963) and Schott (1965): LS - leading shock front, ABCDE - transverse detonation front (spin head), IZ - induction zone; RZ - thin reaction zone behind the lead shock and behind the spin head, AB - the line of triple-shock intersection; (c) the front view of (b). Bold arrows indicate the direction of propagation.

et al. (1959) contains extensive measurements of  $p/d$  made in hydrogen-oxygen mixtures, pure or diluted with argon, nitrogen, or helium at various initial pressures. In near-limit mixtures  $p/d$  was always about three, while in other mixtures it could take on different values, all ranging between about two and six. An example of the mixture with high value of  $p/d$  is the hydrogen-oxygen mixture diluted with helium at high initial pressures.

Low initial pressures of a mixture also favor formation of a steady spin. Generally, at low pressures a single spinning front is observed while an increase in pressure can lead to a formation of more than one rotating fronts. For example, Duff (1961) observed a single-head spin in a stoichiometric hydrogen-oxygen mixture in 5/8 in. diameter tube at initial pressure  $p_0 = 3.99$  mm Hg, while at  $p_0 = 8.49$  mm Hg a diamond-like pattern was observed. As the pressure is increased, the general trend is that the pattern becomes finer and usually very irregular; symmetrical patterns were found to be rather uncommon and unstable. Duff attributes the diamond-shape of the patterns to the interaction of multiple spinning shocks of different helicity, strength, as well as frequency. Duff's investigations also include end-plate soot records which provide certain evidence for the existence of multiple-head spins and their internal structure; the records show that the spin discontinuity extends towards the tube center and often terminates at roughly half the radius from the wall.

Munday, Ubbelohde & Wood (1968) have investigated the effect of dilution on the spin in hydrogen-oxygen mixtures diluted with argon  $2H_2 + O_2 + xAr$ . In a certain range of argon dilution only a single-head spin was observed, while a decrease of argon concentration led to occurrence of a multiple-head spin instead of a single-head spin. Further decreases in the argon content led to a rippled chaotic front.

Manson (1946), Fay (1952), and Chu (1956) independently developed a simple theory which was the best explanation of spin detonation. The basic underlying idea of the theory is the assumption that the spinning detonation front is intimately coupled to the acoustic field in the detonation products. The exact physical mechanism of the coupling is left unexplained, but it is assumed that the frequency of one of the first (typically the first) angular modes of acoustic waves in the tube is equal to the rotation frequency of the spinning front.

The Chapman-Jouguet (CJ) detonation model is assumed with uniform profiles of pressure and temperature (except for acoustic perturbations) behind the CJ plane. The solution of the wave equation in a tube involves three eigenvalues which are determined by the boundary conditions at the wall and at the CJ plane. Periodicity is assumed in the azimuthal direction, and the radial velocity vanishes at the wall. An additional boundary condition which is needed to close the problem is the assumption of a planar transverse vibration at the CJ plane. Although no physical justification is given for the latter assumption, it leads to a formula for the pitch-to-diameter ratio that is in rough agreement with experiments:

$$\frac{p}{d} \approx \frac{\gamma_{prod} + 1}{\gamma_{prod}} \frac{\pi}{k_n}. \quad (1)$$

Here  $k_n$  is the first root of the derivative of the Bessel function of the first kind. For typical values of the adiabatic exponent  $\gamma_{prod}$  in detonation products, the formula gives a value about three for a single-head structure of the spin in close agreement with experimental results. But the theory does not consider the reaction zone which is necessarily involved in mechanism of spinning instability.

In this work we study instability of the one-dimensional steady detonation in a tube subject to three-dimensional linear perturbations by the method of normal modes. The mathematical formulation of the linear instability problem of spinning detonation and the solution technique is similar to the work by Lee & Stewart (1990) used to describe one-dimensional instability analysis of gaseous detonations. One of the main differences in the present work from recent past works is the use of cylindrical coordinates. As we will explain, cylindrical coordinates impose a distinct form of the condition at infinity. Also, the analog to the transverse wave number has discrete modes instead of having continuous variation.

In Section 2 we present the governing equations and the scalings that are used throughout the rest of the paper. Section 3 contains brief discussion of the steady-state one-dimensional solution. In section 4 we present the (linearized) form of the stability equations. In section 5 we derive the radiation condition and discuss the variance of the radiation condition with that found for Cartesian coordinates. In section 6 we present a very brief discussion of the numerical technique used to find eigenvalues, eigenfunctions and neutral stability curves. In section 7 we concentrate on results associated with neutral stability boundaries and how our results are consistent with experimental observations. Section 8 has concluding remarks and section 9 has acknowledgements.

## 2 Governing equations

The governing equations are the Euler equations of gasdynamics for a reactive gas medium, which can be written as

$$\frac{\partial \rho}{\partial t} + \mathbf{u} \cdot \nabla \rho + \rho \nabla \cdot \mathbf{u} = 0, \quad (2)$$

$$\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} + \frac{1}{\rho} \nabla p = 0, \quad (3)$$

$$\frac{\partial h}{\partial t} + \mathbf{u} \cdot \nabla h - \frac{1}{\rho} \frac{\partial p}{\partial t} = 0, \quad (4)$$

$$\frac{\partial \lambda}{\partial t} + \mathbf{u} \cdot \nabla \lambda = \omega. \quad (5)$$

The energy equation (4) is written in terms of the total enthalpy  $h = e + p/\rho + \mathbf{u}^2/2$ , where  $e$  is the specific total energy,  $p$  is the pressure,  $\rho$  is the density,  $\mathbf{u}$  is the velocity,  $\omega$  is the reaction rate and  $\lambda$  is the reaction progress variable. A single progress variable for exothermic reaction is assumed with an Arrhenius simple-depletion form for the reaction rate law and an ideal thermal equation of state is also assumed,

$$e = \frac{1}{\gamma - 1} \frac{p}{\rho} - Q\lambda, \quad \omega = k(1 - \lambda)^\nu \exp(-E/RT), \quad RT = p/\rho. \quad (6)$$

The energy equation can be rewritten in terms of the pressure as

$$\frac{\partial p}{\partial t} + \mathbf{u} \cdot \nabla p + \gamma p \nabla \cdot \mathbf{u} - (\gamma - 1)Q\rho\omega = 0. \quad (7)$$

The normal shock relations for the lead shock must be added to complete the governing equation. We assume that upstream the fluid is motionless with  $\mathbf{u} = 0$ , and use a 0 subscript to label the upstream ambient state and an s-subscript to label the shocked state. Take  $v = 1/\rho$  to be the specific volume. Let  $\mathbf{D} = D\mathbf{n}$  be the normal shock velocity, where  $\mathbf{n}$  is the outward unit normal to the shock. Let unit vectors  $\mathbf{t}_1, \mathbf{t}_2$  lie in the tangent plane of  $\mathbf{n}$ . Then for the ideal equation of state, with the previous assumptions the shock relations can be written as

$$\begin{aligned} \rho(\mathbf{u} - \mathbf{D}) \cdot \mathbf{n}|_0 &= \rho(\mathbf{u} - \mathbf{D}) \cdot \mathbf{n}|_s = m, \quad p - p_0 = m^2(v_0 - v_s), \\ \mathbf{u} \cdot \mathbf{t}_1|_s &= 0, \quad \mathbf{u} \cdot \mathbf{t}_2|_s = 0, \\ \frac{\gamma}{\gamma - 1} \frac{p_0}{\rho_0} + \frac{\mathbf{D}^2}{2} &= \frac{\gamma}{\gamma - 1} \frac{p_s}{\rho_s} + \frac{(\mathbf{u} - \mathbf{D})^2}{2}, \quad \lambda_s = 0. \end{aligned} \quad (8)$$

Next we scale the variables and develop the dimensionless form for governing system. Dimensional scales are based on the one-dimensional steady detonation wave. The density, pressure and velocity scales are steady detonation shock density, pressure and sound speed,  $\rho_s, p_s$  and  $c_s$ . The length scale is chosen to be the steady half reaction zone length,  $\ell_{1/2}$ , i.e. for a chosen steady detonation the length behind the shock for the reaction to reach one half. The time scale is the length scale divided by the velocity scale,  $\ell_{1/2}/c_s$ . To simplify our notation we now use a tilde ( $\sim$ ) to represent dimensional quantities and plain variable to represent dimensionless quantities and an asterisk (\*) to identify quantities of the one-dimensional steady state and  $b$  subscript to refer to the states at the end of the reaction zone.

### 3 The one-dimensional steady state

The one-dimensional steady state is found simply from integrating the conservative form of the governing equations in a frame moving with the detonation. This results in finding the ZND detonation structure. We follow the presentation found in Lee & Stewart (1990) and the results are given again for convenience. If  $u^*$  is the particle velocity in the steady frame, and  $p^*$  and  $\rho^*$  are the steady pressure and density, these are expressed in terms of  $\lambda^*$  by

$$p^* = a + (1 - a)\sqrt{1 - b\beta\lambda^*}, \quad u^* = \frac{1 - p^*}{\gamma M_s} + M_s, \quad \rho^* = \frac{M_s}{u^*}, \quad (9)$$

where  $M_s = u^*/c_s^*$  is the Mach number behind the shock as seen from an observer riding the shock. It is useful to introduce  $D = \tilde{D}^*/\tilde{c}_0$  to represent the steady Mach number of the detonation relative to an observer in ambient material or lab-frame. The constants  $a$  and  $b$ , and the relationship between  $M_s$  and  $D$  are given by

$$a = \frac{1 + \gamma M_s^2}{\gamma + 1}, \quad b = \frac{2\gamma(\gamma - 1)M_s^2}{(\gamma + 1)(1 - a)^2}, \quad M_s^2 = \frac{(\gamma - 1)D^2 + 2}{2\gamma D^2 - (\gamma - 1)}. \quad (10)$$

At the shock front the steady state variables satisfy

$$\rho^* = 1, p^* = 1, u^* = M_s, \quad \text{and} \quad \lambda^* = 0. \quad (11)$$

The dimensionless heat release and the activation energy are defined as  $\beta = \tilde{Q}\gamma/(\tilde{c}_s^*)^2$  and  $\Theta = \tilde{E}\gamma/(\tilde{c}_s^*)^2$  respectively. While it is convenient to carry out the analysis in post shock scales, the effect of the scaling is to make the scales dependent on the reaction zone length and in particular the activation energy. Many results of simulation are reported for activation energies and heat release scale with respect to upstream condition (Erpenbeck's scales). Plain  $E$  is used to represent the scaled activation energy  $E = \gamma\tilde{E}/\tilde{c}_0^2$  and plain  $Q$  is used to represent the scaled heat release  $Q = \gamma\tilde{Q}/\tilde{c}_0^2$ .

The overdrive factor is defined as  $f = (D/D_{CJ})^2$  where  $D_{CJ} = \tilde{D}^*/\tilde{c}_0$  is the scaled Chapman-Jouguet detonation velocity given by the formula

$$D_{CJ} = \sqrt{1 + \frac{(\gamma^2 - 1)Q}{2\gamma}} + \sqrt{\frac{(\gamma^2 - 1)Q}{2\gamma}}. \quad (12)$$

The spatial structure of  $\lambda^*$  is obtained by integration of the rate equation as

$$z = \int_0^{\lambda^*} \frac{u^*(\lambda)}{\omega^*(\lambda)} d\lambda = \frac{1}{k} \int_0^{\lambda^*} \frac{u^*(\lambda)}{(1 - \lambda)^\nu} e^{\Theta/(p^*/\rho^*)} d\lambda. \quad (13)$$

The value of  $k = \tilde{k}\tilde{\ell}_{1/2}^*/\tilde{c}_s^*$  is fixed by setting  $z = 1$  when  $\lambda^* = 1/2$  in the above integral.

## 4 The linear stability problem

Here we present the formulation of the linear stability problem that determines the normal modes. We use cylindrical coordinates attached to the perturbed shock front. The superscript  $l$  below is used to indicate variables  $(r^l, \theta^l, z^l, t^l)$  represented in the laboratory frame. Let  $D_s = \tilde{D}^*/\tilde{c}_s^*$  be the steady detonation shock velocity scaled by the steady post shock sound speed. Next we introduce the shock attached coordinates  $(r, \theta, z, t)$  through the coordinate transformation

$$z = z^l - D_s t^l - \psi(r^l, \theta^l, t^l), \quad r = r^l, \quad \theta = \theta^l, \quad t = t^l. \quad (14)$$

The function  $\psi$  represents the shock front displacement from the unperturbed position. The instantaneous location of the shock front is at  $z = 0$  in the shock-attached coordinates. Note that the velocity of the shock attached frame is  $D_s + \partial\psi/\partial t$ , hence the particle velocity in the frame is  $u_z = u_z^l - D_s - \partial\psi/\partial t$ . This choice is consistent with the definition of  $u_z$  found in the detonation shock dynamics studies that use intrinsic shock-attached coordinates, where the shock acceleration of the frame explicitly appears in the governing equations, see for example, Yao & Stewart (1996). In previous linear stability works such as Lee & Stewart (1990) and Short & Stewart (1998), the particle velocity in the steady frame,  $u_z^l - D_s$  (not the  $z$  velocity component in the shock attached frame) is used instead as the dependent  $z$  velocity component, which follows Erpenbeck's earlier precedent. The formulation here differs slightly, and the shock relations, in particular are affected. However either choice could have been made without any effect on the conclusions.

If we introduce the operators  $\Phi$  and  $L$  defined by

$$\Phi \equiv u_r \frac{\partial\psi}{\partial r} + \frac{u_\theta}{r} \frac{\partial\psi}{\partial\theta}, \quad L \equiv u_r \frac{\partial}{\partial r} + \frac{\partial u_\theta}{\partial r} \frac{\partial}{\partial\theta} + (u_z - \Phi) \frac{\partial}{\partial z}, \quad (15)$$

then the transformed set of the governing equations becomes

$$\frac{\partial\rho}{\partial t} + \mathbf{u} \cdot \nabla\rho + \rho \nabla \cdot \mathbf{u} - \frac{\partial\rho\Phi}{\partial z} = 0, \quad (16)$$

$$\frac{\partial u_r}{\partial t} + L(u_r) - \frac{u_\theta^2}{r} + \frac{1}{\gamma\rho} \frac{\partial p}{\partial r} - \frac{1}{\gamma\rho} \frac{\partial\psi}{\partial r} \frac{\partial p}{\partial z} = 0, \quad (17)$$

$$\frac{\partial u_\theta}{\partial t} + L(u_\theta) + \frac{u_r u_\theta}{r} + \frac{1}{\gamma\rho r} \frac{\partial p}{\partial\theta} - \frac{1}{\gamma\rho r} \frac{\partial\psi}{\partial\theta} \frac{\partial p}{\partial z} = 0, \quad (18)$$

$$\frac{\partial u_z}{\partial t} + L(u_z) + \frac{1}{\gamma\rho} \frac{\partial p}{\partial z} + \frac{\partial^2\psi}{\partial t^2} = 0, \quad (19)$$

$$\frac{\partial p}{\partial t} + L(p) + \gamma p \nabla \cdot \mathbf{u} - (\gamma - 1)\beta \rho r - \gamma p \frac{\partial \Phi}{\partial z} = 0, \quad (20)$$

$$\frac{\partial \lambda}{\partial t} + L(\lambda) = \omega. \quad (21)$$

We note that  $\mathbf{u} = (u_r, u_\theta, u_z)$ , and that in cylindrical polar coordinates,  $\mathbf{u} \cdot \nabla = u_r \partial / \partial r + (u_\theta / r) \partial / \partial \theta + u_z \partial / \partial z$  and  $\nabla \cdot \mathbf{u} = \partial u_r / \partial r + u_r / r + (1/r) \partial u_\theta / \partial \theta + \partial u_z / \partial z$ .

The governing equations can also be written in a matrix form. Let the column vector  $\mathbf{q} = [\rho, u_r, u_\theta, u_z, p, \lambda]^T$ . Then the equations in the shock-attached frame can be recast as

$$\frac{\partial \mathbf{q}}{\partial t} + \mathbf{A}_z \frac{\partial \mathbf{q}}{\partial z} + \mathbf{A}_r \frac{\partial \mathbf{q}}{\partial r} + \mathbf{A}_\theta \frac{1}{r} \frac{\partial \mathbf{q}}{\partial \theta} + \frac{\mathbf{a}}{r} - \mathbf{B}_r \frac{\partial \mathbf{q}}{\partial z} \frac{\partial \psi}{\partial r} - \mathbf{B}_\theta \frac{\partial \mathbf{q}}{\partial z} \frac{1}{r} \frac{\partial \psi}{\partial \theta} + \mathbf{b} \frac{\partial^2 \psi}{\partial t^2} = \mathbf{c}, \quad (22)$$

where the matrices and column vectors are written out in Appendix I. If the equations were steady and plane, then the governing equations are simply  $\mathbf{A}_z \partial \mathbf{q} / \partial z = \mathbf{c}$ .

The equations are expanded about the steady-state solution and we seek solutions to the normal mode of the form

$$\mathbf{q} = \mathbf{q}^*(z) + \mathbf{q}' \exp(\alpha t + i n \theta), \quad \psi = \psi' \exp(\alpha t + i n \theta), \quad (23)$$

where the prime superscript implies a small amplitude in a norm associated with deviation of the initial conditions from those of plane steady detonation. A straightforward expansion of the various terms found in (22) combined with the fact that the steady state only depends on  $z$  leads to

$$\begin{aligned} & \alpha \mathbf{q}' + \mathbf{A}_z^* \frac{\partial \mathbf{q}'}{\partial z} + \mathbf{A}_r^* \frac{\partial \mathbf{q}'}{\partial r} + i n \mathbf{A}_\theta^* \frac{1}{r} \mathbf{q}' + \mathbf{A}_z' \frac{\partial \mathbf{q}^*}{\partial z} \\ & + \frac{\mathbf{a}'}{r} - \mathbf{B}_r^* \frac{\partial \mathbf{q}^*}{\partial z} \frac{\partial \psi'}{\partial r} - i n \mathbf{B}_\theta^* \frac{\partial \mathbf{q}^*}{\partial z} \frac{1}{r} \psi' + \alpha^2 \mathbf{b}^* \psi' = \mathbf{c}'. \end{aligned} \quad (24)$$

Since all the perturbations are uniformly the same order ( $O(\epsilon)$  say) we can now consider the primed variables to be the  $O(1)$  coefficient functions.

Then the normal modes are separable in  $r, \theta, z$  by writing

$$\mathbf{q}' = \begin{bmatrix} \rho'(z) J_n \\ u_r'(z) dJ_n/dr \\ u_\theta'(z) J_n/r \\ u_z'(z) J_n \\ p'(z) J_n \\ \lambda'(z) J_n \end{bmatrix} \quad \text{and} \quad \psi' = J_n. \quad (25)$$



Here  $J_n = J_n(kr)$  is the Bessel function of the first kind of integer index  $n$ ,  $k$  is the radial wavenumber to be determined from the boundary condition at the wall and  $\alpha$  is a complex growth rate of perturbations.

It follows from the above form of the normal modes that the  $u_r$  and  $u_\theta$  equations yield the relationship

$$u'_\theta - i n u'_r = C \exp\left(-\alpha \int_0^z \frac{dz}{u_z^*}\right)$$

where the constant  $C$  when found from RH conditions (which imply  $u'_\theta(0) = i n u'_r(0)$ ) turns out to be zero. Thus

$$u'_\theta = i n u'_r \quad (26)$$

and once the  $u'_r$  perturbation is found,  $u'_\theta$  follows directly. This reduces the number of complex ODEs from six to five. Without loss of generality we can also take the shock displacement perturbation amplitude to be normalized to one since the eigenfunctions are determined up to a multiplicative constant.

The stability equations can be re-written in terms of a new column vector,  $\mathbf{f}' = [\rho', u'_r, u'_z, p', \lambda']^T$ , representing the complex perturbations, as follows

$$\alpha \mathbf{f}' + \mathbf{A}^* \frac{d\mathbf{f}'}{dz} + \mathbf{C}^* \mathbf{f}' - \mathbf{b}^* = 0, \quad (27)$$

where  $\mathbf{A}^*$ ,  $\mathbf{C}^*$ , and  $\mathbf{b}^*$  are given by

$$\mathbf{A}^* = \begin{bmatrix} u_z & 0 & \rho & 0 & 0 \\ 0 & u_z & 0 & 0 & 0 \\ 0 & 0 & u_z & 1/(\gamma\rho) & 0 \\ 0 & 0 & \gamma p & u_z & 0 \\ 0 & 0 & 0 & 0 & u_z \end{bmatrix}^*, \quad \mathbf{b}^* = \begin{bmatrix} 0 \\ -p_{,z}/(\gamma\rho) \\ \alpha^2 \\ 0 \\ 0 \end{bmatrix}^*, \quad (28)$$

$$\mathbf{C}^* = \begin{bmatrix} u_{z,z} & -k^2\rho & \rho_{,z} & 0 & 0 \\ 0 & 0 & 0 & 1/(\gamma\rho) & 0 \\ -p_{,z}/(\gamma\rho^2) & 0 & u_{z,z} & 0 & 0 \\ -(\gamma-1)\beta(\omega + \rho\omega_{,\rho}) & -\gamma k^2 p & p_{,z} & \gamma u_{z,z} - (\gamma-1)\beta\rho\omega_{,p} & -(\gamma-1)\beta\rho\omega_{,\lambda} \\ -\omega_{,\rho} & 0 & \lambda_{,z} & -\omega_{,p} & -\omega_{,\lambda} \end{bmatrix}^*. \quad (29)$$

Note that  $\mathbf{A}^*$  follows directly from the definition of  $\mathbf{A}_z^*$ . The matrix  $\mathbf{C}^*$  has contributions from all the other  $\mathbf{A}^*$  terms. The term  $\mathbf{b}^*$  is determined from the shock displacement. The reaction rate sensitivities are defined by the expansion of the reaction rate as  $\omega' = \omega_{,p}^* p' + \omega_{,\rho}^* \rho' + \omega_{,\lambda}^* \lambda'$ , where  $\omega_{,p}^* = \Theta \rho^* \omega^* / (p^*)^2$ ,  $\omega_{,\rho}^* = -\Theta \omega^* / p^*$ , and  $\omega_{,\lambda}^* = -\nu \omega^* / (1 - \lambda^*)$ . Notice, that if  $\nu \geq 1$  there is no singularity in the system at any point, except for the CJ

case, for which one of the eigenvalues of  $\mathbf{A}^*$  becomes zero at infinity. The CJ case is treated by a limit as the overdrive factor  $f$  approaches one.

The Rankine-Hugoniot relations (8) are expanded to obtain the conditions at the shock, which is located at  $z = 0$ . Their linearization (with  $\psi' = 1$ ) leads to

$$\begin{aligned} \rho' &= -\frac{4}{(\gamma + 1)D^2 M_s} \alpha, & u_z' &= \frac{2 - (\gamma - 1)D^2}{(\gamma + 1)D^2} \alpha, & u_r' &= -\frac{1}{M_s} \frac{M_s^2 - D^2}{1 + \gamma D^2} \alpha, \\ p' &= -\frac{4\gamma M_s}{\gamma + 1} \alpha, & \lambda' &= 0. \end{aligned} \quad (30)$$

The boundary condition at the tube wall is that the radial velocity is zero,  $u_r = 0$ . It follows then that

$$\left. \frac{dJ_n(kr)}{dr} \right|_{r=a} = 0. \quad (31)$$

Equation (31) determines all possible wave numbers  $k_{nm}$  for the integer  $n = 0, 1, 2, \dots$  and  $m = 1, 2, \dots$ . In what follows,  $n$  will be identified with the number of spin heads, while  $m$  is the radial mode number such that  $m - 1$  gives the number of zeroes of the Bessel function on  $0 < r < a$ . Note that the eigenfunctions given by  $J_0(kr)$  correspond to axisymmetric disturbances with  $u_\theta' = 0$  and  $\partial/\partial\theta = 0$ . An important special case is of the one-dimensional disturbance corresponding to  $k = 0$ . It satisfies the wall boundary condition at all  $a$ , since  $J_0'(0) = 0$ . Thus our normal modes include not only spinning modes, but also axisymmetric non-spinning as well as purely longitudinal modes.

## 5 The radiation condition

One more condition is required to close the above system of ODEs and derive the dispersion relation. It is derived from the condition that the initial perturbations are uniformly bounded in space. Spatial unboundedness of unstable eigenfunctions can result from the behavior of the solution at  $z \rightarrow -\infty$ . Hence the necessary condition is identified by considering the linearized system in the rear far field where the steady flow is constant and the reaction is complete. An alternate interpretation of the radiation condition is that the intrinsic instability of the detonation is a result of interaction between the leading shock and the reaction zone only and should not be affected by disturbances that travel towards the shock from an infinite distance behind the reaction zone. Hence it is assumed that the disturbance eigenfunctions are missing the incoming wave family at the end of the reaction zone. In one form or another, such condition has been used by many researchers investigating stability of detonation waves (e.g. Erpenbeck 1962, Buckmaster & Ludford 1986, Lee & Stewart 1990). The radiation condition has a different form in cylindrical coordinates than in Cartesian coordinates, but it expresses the same physical condition. Note that it is possible to apply different conditions, such as for example, the piston condition  $u' = 0$  at infinity, but it should be realized that as a result, the instability spectrum would be altered

by the extra interactions of the piston and leading shock compared to that of the intrinsic mechanism.

Next, we turn to the derivation of the radiation condition in cylindrical coordinates. At the end of the reaction zone where the steady solution reaches a constant state we can set  $\lambda' = 0$  because of the reaction rate being negligible and  $\lambda'$  decaying exponentially with increasing  $z$ . This assumption does not change the overall result and in Appendix II, we derive the form of the radiation condition where  $\lambda'$  is retained. The acoustic solution in the far-field has the same radial dependencies as in the normal mode representation in the main flow. But the dependence on time and the axial coordinate are kept arbitrary.

With these assumptions the acoustic expansion is of the general form  $\mathbf{q} = \mathbf{q}_b + \hat{\mathbf{q}}$  where the hat refers to an acoustic perturbation. The  $u_r, u_z$  and  $p$  equations define the acoustic modes and are given by

$$\mathcal{L}\hat{u}_r + \frac{\hat{p}}{\gamma\rho_b} = 0, \quad (32)$$

$$\mathcal{L}\hat{u}_z + \frac{1}{\gamma\rho_b} \frac{\partial \hat{p}}{\partial z} = 0, \quad (33)$$

$$\mathcal{L}\hat{p} + \gamma p_b \left( \frac{\partial \hat{u}_z}{\partial z} - k^2 \hat{u}_r \right) = 0, \quad (34)$$

where  $\mathcal{L} \equiv \partial/\partial t + u_b \partial/\partial z$  and the subscript  $b$  denotes the burnt state at  $z \rightarrow -\infty$ . (Equation (33) should have the term  $\partial^2 \psi / \partial t^2$  on the left-hand side, but since  $\partial^2 \psi / \partial t^2 = \mathcal{L}(\partial \psi / \partial t)$ , that term is temporarily absorbed into  $\hat{u}_z$ ). If we apply the operator  $\mathcal{L}$  on the equation (34) and make use of equations (32) and (33), then we can get a single equation in  $\hat{p}$ :

$$\frac{\partial^2 \hat{p}}{\partial t^2} + 2u_b \frac{\partial^2 \hat{p}}{\partial t \partial z} - (c_b^2 - u_b^2) \frac{\partial^2 \hat{p}}{\partial z^2} + (c_b k)^2 \hat{p} = 0. \quad (35)$$

Note that this is a Klein-Gordon equation, solutions of which represent dispersive travelling waves. In terms of the characteristic variables  $\xi = z - (u_b + c_b)t$  and  $\eta = z - (u_b - c_b)t$  it can also be written as  $\partial^2 \hat{p} / \partial \xi \partial \eta - (k^2/4) \hat{p} = 0$ .

We look for a solution of equation (35) which is of the travelling wave form  $\exp(\alpha t + s z)$ . Substituting  $\hat{p} = \exp(\alpha t + s z)$  into equation (35) we get a dispersion relation for the complex axial wavenumber  $s(\alpha)$

$$\alpha^2 + 2u_b \alpha s - (c_b^2 - u_b^2) s^2 + c_b^2 k^2 = 0, \quad (36)$$

which has roots given by

$$s = \frac{1}{c_b^2 - u_b^2} \left( \alpha u_b + c_b [\alpha^2 + k^2 (c_b^2 - u_b^2)]^{1/2} \right). \quad (37)$$

Note that in the Chapman-Jouguet limit,  $f = 1$  and  $D = D_{CJ}$  and it follows that the flow state at the end of the reaction zone is sonic with  $c_b = -u_b$ , and (37) reduces to

$$s = -\frac{1}{2u_b} \left( \alpha + \frac{c_b^2 k^2}{\alpha} \right). \quad (38)$$

The far-field dispersion relation has two branches of the square root so that the unstable disturbances with  $Re(\alpha)$  are spatially bounded only if the branch with positive real part is kept. From the traveling-wave character of solutions to the Klein-Gordon equation it is clear that the boundedness requirement is equivalent to elimination of acoustic waves propagating from  $z \rightarrow -\infty$  towards the shock.

It is worth pointing out that the situation is not different in principle if we consider the case with a finite reaction zone. Even though the reaction zone does not extend to infinity, and the spatial unboundedness is of no concern anymore, the radiation condition is imposed based on the physics of the problem: the development of unstable perturbations has to occur only due to the interaction between the lead shock front and the reaction zone. The boundary value problem should now be set up for a finite domain and the radiation condition be applied at the end of the reaction zone.

Equation (34) can be rearranged as

$$k^2 \hat{u}_r = \frac{\alpha + u_b s}{\gamma p_b} \hat{p} + \frac{\partial \hat{u}_z}{\partial z}. \quad (39)$$

The time dependence of all the variables has to be the same as in (25), i.e. exponential. The axial dependence is known only upon the solution of the exact normal mode system, but it has to satisfy equation (39). Now, writing  $s$  in terms of  $\alpha$  from (37) and plugging  $\hat{p} = p'(z)exp(\alpha t)$ ,  $\hat{u}_z = u'_z(z)exp(\alpha t)$ ,  $\hat{u}_r = u'_r(z)exp(\alpha t)$  into (39) we get the *radiation condition*

$$\frac{du'_z}{dz} - k^2 u'_r + \frac{\alpha c_b + u_b[\alpha^2 + k^2(c_b^2 - u_b^2)]^{1/2}}{\gamma \rho_b c_b (c_b^2 - u_b^2)} p' = 0. \quad (40)$$

By choosing the positive branch of the square root in this equation we explicitly eliminate incoming waves at infinity. Note that for CJ detonation ( $c_b/(-u_b) = 1$ ) the radiation condition becomes

$$\frac{du'_z}{dz} - k^2 u'_r + \frac{\alpha - k^2 c_b^2 / \alpha}{2\gamma \rho_b c_b^2} p' = 0. \quad (41)$$

Also, for the one-dimensional case letting  $k \rightarrow 0$  we integrate (40) and noting that  $\int p' dz = const \int exp(sz) dz = p'/s$ , we obtain the one-dimensional radiation condition given, for example, in Lee & Stewart [14] with  $\lambda'$  set to 0,

$$u'_z + \alpha + \frac{1}{\gamma \rho_b c_b} p' = 0. \quad (42)$$

The extra term  $\alpha$  in this equation is due to the definition of the axial velocity which is different from that in Lee & Stewart [14], i.e. if  $u_z^{LS}$  denotes the axial velocity in Lee & Stewart [14], then  $u_z^{LS} = u_z + \partial\psi/\partial t$  and  $\alpha$  in (42) comes from  $\partial\psi/\partial t$ .

## 6 The numerical solution and technique for finding roots

The unstable eigenvalues are found by solving the radiation condition (40) numerically by means of DNSQE subroutine from NIST mathematical software repository (2001). The subroutine solves a system of nonlinear equations by the Powell hybrid method, a version of the Newton method. To provide the subroutine with an initial guess we first determine the approximate location of the roots of (40) by plotting the magnitude of the left-hand side of (40) (lets denote it  $H$ ) in some bounded domain of the complex plane  $\alpha$ . This is the “carpet search” method, first introduced by Lee & Stewart (1990) to find the eigenvalues for the one-dimensional detonation. The surface of  $|H|$  has clear domains of attraction at the locations of the roots, thus allowing one to obtain the guess from the contour plot of  $|H|$ . Once the guess is supplied to DNSQE, the subroutine then finds the exact value within a given tolerance (typically  $10^{-6} - 10^{-8}$ ) iteratively. At each iteration the entire solution of the ODE’s (27) is required. The latter is obtained by means of DDASSL subroutine developed by L. Petzold, which can be found at NIST (2001). This subroutine is based on Gear’s method for stiff differential-algebraic systems. The solution for the ODE’s is again obtained within a given tolerance, which is typically  $10^{-8} - 10^{-10}$ . Such accuracy is necessary in order to achieve proper convergence for the root solver. This is because of the property of the radiation condition that the domains of attraction of the roots are extremely small, especially at high frequencies; thus large steps during iterations can keep the solver wandering around the root or make it leave the domain entirely. The efficiency of DNSQE and DDASSL for a similar class of problems was demonstrated by Short & Stewart (1998).

The neutral stability curves are constructed using the arclength continuation method. In this method each subsequent point on the curve is found over a fixed length along the curve. This method avoids numerical difficulties associated with turning points.

## 7 Discussion of results for spinning instability

Low-frequency spin (i.e. with small number of rotating fronts) is typically observed in experiments with explosive mixtures close to their detonability limits (usually lean limits) so that the heat effect  $Q$  is small compared to that of stoichiometric composition. This is the main reason for our focus on the range of  $Q$  between 0 and 20 in the following calculations. Small  $Q$  implies that the detonation speed and therefore post-shock temperature is small. Strong dependence of the reaction rate on the post-shock temperature leads to a significant increase in the induction zone length. As the degree of dilution decreases, the number of spin heads is observed to grow. For example, in hydrogen-oxygen-argon mixtures Munday et al. (1968) found that single-head spin exists within the range of dilution between 88.8 and 91.9 volume percent of argon, while in the range between 87.0 and 88.8 percent, the four-head spin was observed, bifurcating from a single-head mode at 88.8 percent argon. Two- and three-head spins were not observed.

m\ n	1	2	3	4	5	6	7
1	1.841	3.054	4.201	5.318	6.416	7.501	8.578
2	5.331	6.706	8.015	9.282	10.520	11.735	12.932
3	8.536	9.969	11.346	12.682	13.987		

Table 1: First several roots of  $J'_n(x_{nm}) = 0$ .

We next discuss some of the results of the stability analysis and try to relate them to the above-mentioned experimental findings. Note that the set of bifurcation parameters is comprised of the heat release  $Q$ , activation energy  $E$ , overdrive factor  $f$ , adiabatic exponent  $\gamma$ , and the radial wavenumber  $k$  (recall that  $k \equiv 2x_{nm}/d$ , where  $x_{nm}$  is the  $m - th$  root of the Bessel function's derivative,  $J'_n(x_{nm}) = 0$ ,  $d$  is the tube diameter,  $n$  is the number of maxima in the angular direction (number of spinning heads),  $m - 1$  gives the number of nodes of  $J_n(kr)$  along the radius; thus  $2\pi/k$  is the characteristic wavelength in the radial direction). In this paper we do not attempt to cover the entire space of bifurcation parameters, but rather focus on the detailed analysis of some of them, choosing others on the basis of experimental knowledge about spin detonation. In particular, we keep  $f$  fixed at  $f = 1.0$ , since the experiment indicates that overdrive tends to suppress spin detonation. Also, we look at only two values of the adiabatic exponent,  $\gamma = 1.3$  and  $\gamma = 1.6$  and two values of the tube diameter,  $d = 2$  and  $d = 20$  (recall that the length scale is the half-reaction length  $\tilde{l}_{1/2}$ , so that these tube diameters are twice and twenty times the length of the reaction zone, respectively). We do vary  $Q$  and  $E$  since  $Q$  is related to the degree of dilution (the higher the degree of dilution the smaller the heat effect  $Q$ ), and different  $E$ 's represent mixtures of different reactivity. We vary  $n$  and  $m$  since these provide a selection of particular modes of spin and, which is very important for the link between the instability results and experiment, gives us the spin pitch to diameter ratio  $p/d$  for the most unstable mode. Finally, we calculate the dependence of the neutrally stable roots on the radial wave number  $k$ . This lets us avoid computing the huge number of discrete modes, labeled with three indices,  $n$ ,  $m$ , running through one to infinity each, as well as the mode index  $j$  in the temporal spectrum, which may be as large as tens or even hundreds.

First, let us consider the case with fixed values of  $d = 20$ ,  $\gamma = 1.6$ , and  $f = 1.0$ . These choices are motivated by the following facts from experiments as well as our own calculations on one-dimensional detonation instability. Experiments show that typical cell size of cellular detonation is of the order of 10 to 100 reaction-zone lengths (see, e.g. Fickett & Davis 2001), so that we expect the radial wavenumber for the most unstable mode to be around  $0.06 - 0.6$ . For  $d = 20$ , the smallest wavenumber is  $k = 2x_{11}/d = 0.184$  (see Table 1).

The choice of  $\gamma = 1.6$  is motivated, first, by the experimental results from Gordon et al. (1959) in which the experiments were carried out mostly in hydrogen-oxygen mixtures diluted with monatomic inert gases, such as helium and argon, which render large  $\gamma$  for the mixture (pure helium or argon have  $\gamma = 1.67$ ); secondly, our calculations of one-dimensional instability for various  $\gamma$  indicate that larger  $\gamma$  tends to strongly stabilize detonations with larger  $Q$  while destabilizing, albeit insignificantly, detonations at smaller  $Q$ , as shown in figure 2. The remaining choice, for  $f = 1.0$  is motivated again by experimental reports such as those in Gordon et al. (1959), which show that the emergence of spin detonation is most

frequent at CJ velocities. This limit is achieved by computing for values of  $f$  close, but not exactly equal to one, namely  $f = 1.01$ . (In fact, the limit  $f \rightarrow 1$  is well behaved in the sense that the solution for the governing equations converges to a limit, although extremely slowly as  $f$  gets closer to one. There is no appreciable difference in the results between  $f = 1.01$  and say  $f = 1.0001$ , but the latter is prohibitively time-consuming computationally.) Also, we did carry out calculations for  $\gamma = 1.3$  and  $d = 2$ , however in less detail than for the case with  $\gamma = 1.6$  and  $d = 20$ , and they reveal some additional features of detonation instability which we shall discuss below.

Figures 3(a) and 3(b) show the neutral stability curves for several spinning modes, ranging from  $n = m = 1$  to  $n = 10, m = 1$  which corresponds to the range of the radial wavenumber  $k$  from  $k = 0.184$  to  $k = 1.189$ . In figure 3(a), the leftmost boundary (the left solid line) is comprised of three neutral stability curves, namely  $n = 3, m = 1$  below  $Q = 16.1$  and  $n = 4, m = 1$  or  $n = 1, m = 2$  above  $Q = 16.1$ . Similarly, in figure 3(b) the leftmost boundary consists of  $n = 3, m = 1$  below  $Q = 0.69$  (dashed line),  $n = 4, m = 1$  or  $n = 1, m = 2$  from  $Q = 0.69$  to  $Q = 8.5$ , and  $n = 5, m = 1$  from  $Q = 8.5$  to  $Q = 20$ . The thick solid line on both figures is the one-dimensional neutral stability boundary corresponding to  $k = 0$ . The horizontal dashes indicate the bifurcation points. The dotted lines in both figures correspond to several neighboring modes below and above the ones which comprise the leftmost boundary. To avoid cluttering we show only those curves which are in the neighborhood of the leftmost boundary.

The main features of figures 3(a) and 3(b) are, first, that there exist spinning modes which are more unstable than (are to the left of) one-dimensional modes in the entire range of  $Q$  and  $E$  under investigation, and second, there exists a leftmost mode, perhaps different for different  $Q$ , such that all modes below and above it are less unstable. As we change  $Q$ , we may encounter bifurcations between different modes as indicated by horizontal dashes on figure 3. In addition, note that many neutral stability curves are not far apart from the leftmost boundary, i.e. corresponding unstable modes have a growth rate very close to the largest one. In fact, the larger the tube diameter the closer the neutral stability curves are to each other and many different modes will have growth rates near the maximum. In contrast, if the tube diameter is small, but still large enough so that spinning instability overtakes one-dimensional instability (see below), then the bifurcations are more pronounced.

Figure 4 shows the steady state flow profiles and neutrally stable eigenfunctions corresponding to two leftmost modes  $n = 3, m = 1$  and  $n = 4, m = 1$  at the bifurcation value of  $Q = 0.69$  and  $E = 23.23$  on figure 3(b). The temporal frequencies of the two modes are  $Im(\alpha) = 0.3798$  for  $n = 3$  and  $Im(\alpha) = 0.4499$  for  $n = 4$ . It is seen that there is little qualitative change in the behavior of the eigenfunctions for the two modes, except the amplitudes are somewhat larger for  $n = 4$  mode. The eigenfunctions shown on figures 4(a) and 4(b) are in fact only the axial amplitudes. The corresponding angular and radial parts bring up a significant change in the three-dimensional shape of the perturbation:  $n = 3$  represents a wave with three crests in the azimuthal direction while there are four crests for the case with  $n = 4$ .

In order to find out the behavior of neutral stability curves for higher modes (hence larger  $k$ 's), we trace the neutrally stable roots as  $k$  is varied at fixed  $Q$ . For example, we pick a value of  $Q = 10$  on figure 3(a) and starting from the one-dimensional neutral stability curve ( $k = 0$ ) calculate the dependence of  $E$  on  $k$ . The result is shown in figure 5(a). Figure 5(b)

shows similar curves corresponding to figure 3(b). The numbering of the curves corresponds to the order in the temporal spectrum, i.e. 1 is the lowest frequency root, 2 is the next and so on. Several important conclusions can be drawn from these figures. First of all, the low  $k$  behavior which is shown in figures 3(a) and 3(b) with a distinct leftmost neutral stability boundary, terminates for the case with  $\gamma = 1.3$  due to the mode  $j = 2$  becoming more unstable at much larger  $k$ , and hence much larger  $n$  and  $m$ . This means that at  $Q = 10$  and  $\gamma = 1.3$  we have high-frequency instability dominant. This may be the reason for the experimental observation, that when the mixture becomes more energetic (less dilute), the low-frequency spin detonation turns into a detonation with a “rippled irregular front” (e.g. Munday et al. 1968, Duff 1961). (The peculiar behavior of the second root in figure 5(a) and third root in figure 5(b) which exhibit loops is indicative of the complicated mathematical structure of the dispersion relation and is not of a numerical origin - hundreds of points are calculated along the loops alone, with tolerances on the order of  $10^{-8} - 10^{-9}$ ). Note, that there is a region of smaller  $Q$  for which low-frequency instability prevails as figures 6(a) and 6(b) show. Thus at sufficiently small  $Q$  there is no high-frequency instability.

Since the linear analysis predicts that large number of unstable modes can have essentially same growth rate if the tube diameter  $d \gg 1$  and it becomes hard to make a selection of a particular mode, it is important to study the nonlinear evolution of the perturbations to capture the experimentally observed behavior of detonation waves and be able to predict the dynamics of the waves more conclusively. It is especially important in view of the very high frequency (large  $k$ ) instability at large  $Q$  and small  $\gamma$  predicted by the linear analysis.

The case with  $\gamma = 1.6$  does not show the high-frequency instability, at least in the reasonable range of  $k$ . (The reader should realize that this work can offer no guarantee that at much larger  $k$  we do not get again the high-frequency mode being the leftmost boundary, especially in view of the peculiarities of the dispersion relation. The reason is that it is prohibitively expensive to compute large  $k$  limit, because the eigenfunctions become highly oscillatory - their frequency  $Im(\alpha_i)$  grows linearly with  $k$  as  $k \rightarrow \infty$ . An asymptotic solution, similar to that of Erpenbeck 1966, would in principle answer the question, but the analysis is rather involved and has yet to be satisfactorily resolved even for the simpler two-dimensional planar case.) Once the neutral stability boundary corresponds to the turning point for root 1 in figure 5(a), it means that there exist a specific wave shape, with specific  $k = k_c$  (inverse of the radial wave length),  $n_c$  (number of spinning heads), and  $m_c$ , which has the maximum growth rate. From figure 5(a) we find  $k_c = 0.46$ , and from figure 3(a)  $n_c = 3$ , and  $m_c = 1$ .

Further support for the results having direct relevance to spin detonation is furnished by figure 7 which shows how the spin pitch to diameter ratio varies along the neutral stability curves in figure 3(a) for the modes with  $n = 3$ ,  $m = 1$  and  $n = 4$ ,  $m = 1$ , which are most unstable modes within the range of  $Q$  from 0 to 20. The ratio is calculated based on the temporal frequency  $\alpha_i = Im(\alpha)$  of a neutrally stable root as

$$\frac{p}{d} = \frac{2\pi n D_s^*}{\alpha_i d}.$$

The predicted values are in good agreement with experiment with  $p/d$  being close to three for a low-frequency spin and being generally about three to six (cf. Gordon, et al. [13]).

The values of  $p/d$  measured in experiments are for detonation with larger (nonlinear)



shock displacement than can be predicted simply from linear stability theory. And directly relating the frequencies of the *linear* perturbations to those of the actual nonlinear spin detonation is not entirely correct and the comparison is more properly regarded as an order of magnitude comparison. This caveat made, it is also worth pointing out that the pitch ratio is essentially determined by the imaginary part of the complex growth rate  $\alpha_i$ . Research that dates back to Erpenbeck's original calculations of the frequency of unstable one-dimensional (galloping) detonation and comparison with Fickett and Wood's direct simulation, see Fickett & Davis (2001), have shown that the frequency of the instability from stability theory is often maintained as a feature of the large amplitude manifestation of the instability. Many researchers have shown this subsequently in the past ten years (see e.g. Short & Quirk 1997).

Figure 8 shows the case of a small tube diameter  $d = 2$  and  $\gamma = 1.3$ . First of all, note that the smallest radial wavenumber is now  $k = 2x_{11}/d = 1.841$  and according to figure 5(b) there exists no part of the curve below  $k = 1.841$  (except a single point at  $k = 0$ ) and hence no turning point region. That is why at sufficiently small  $Q$  the one-dimensional instability dominates, while at higher  $Q$  the high-frequency modes are more unstable. This result indicates that detonation of sufficiently dilute mixtures (small  $Q$ ) in sufficiently small tubes would exhibit pulsating instability and thus lead to galloping detonation. This is also in agreement with experiment, see for example, Voitsekhovskii et al. (1963), Strehlow (1978), Vasiliev (1991), Fickett & Davis (2001).

Let us now discuss what happens if we change various bifurcation parameters. To begin with, consider the limit of a large tube diameter  $d \rightarrow \infty$ . In this limit the distance between neighboring roots  $x_{nm}$  goes to zero, and hence we get a near-continuous variation of  $k$ . Still, figure 5(a) is in force with its turning point. Thus, no matter what the diameter is (if sufficiently large), there exists a characteristic radial wavenumber  $k_c$  (e.g.  $k_c \approx 0.46$  at  $Q = 5$ ) for which the perturbation is most unstable. This corresponds to a certain pair of numbers  $n, m$  dependent on  $d$  that can be found from  $x_{nm} = k_c d/2$ , however we emphasize that the wavelength in the radial direction becomes *independent* of  $d$  as  $d \rightarrow \infty$ , as it clearly should. At large  $k_c r$  the Bessel function  $J_n(k_c r)$  can be approximated by its asymptotic form  $J_n(k_c r) \sim \sqrt{2/(\pi k_c r)} \cos(k_c r - \pi n/2 - \pi/4)$ , from which it is clear that the characteristic wavelength of the most unstable mode is  $\lambda_c = 2\pi/k_c \approx 13.7$ . Thus, we conclude that the transverse cell size based on the linear stability analysis is about 14 times the size of the reaction zone at these particular values of  $Q = 5$  and  $\gamma = 1.6$ . In general, we expect  $k_c$  to be a function of  $Q$  and  $\gamma$ , and thus we can write that

$$\tilde{\lambda}_c = \frac{2\pi}{k_c(Q, \gamma)} \tilde{l}_{1/2}.$$

The value of  $k_c(Q, \gamma)$  can be determined from the stability analysis,  $\tilde{l}_{1/2}$  is determined by the kinetics and the thermodynamic properties of the mixture.

It is interesting to compare the wavelength predicted by the above formula against experiment and numerical calculations. If we take the data from the detonation database at Caltech (2001) and the steady reaction-zone length calculated using detailed chemical mechanism, we have the following data for  $2H_2 + O_2 + 26Ar$  (90%  $Ar$ ): in the fresh mixture  $\gamma_0 = 1.64$ , in the von Neumann state  $\gamma_{vN} = 1.6$ , effective heat release and activation energy are  $Q \approx 7$  and  $E \approx 34$ , respectively. The experimental value of the cell width  $\lambda_w$  is about

40 mm, and the length of the reaction zone  $l_{rz}$  (based on maximum temperature gradient) is 1.23 mm. Their ratio gives  $\lambda_w/l_{rz} = 33$ . The difference of a factor of 2 can be attributed to the ambiguities in the definitions for various quantities. For example, in our steady-state model the position of  $\lambda = 1/2$  is not necessarily at the same point where the temperature gradient attains a maximum. Ambiguities in the definitions of  $E$  and  $\gamma$  can further influence the result. This comparison should be regarded as an order of magnitude comparison, and in that sense the agreement is good. More accurate and careful comparison can be made.

Dilution affects the detonation stability most significantly through its relationship to the heat release  $Q$  and  $\gamma$ . If the diluent is a monatomic gas then we expect large values for  $\gamma$ . Lighter diluents such as helium would also increase the sound speed  $c_0$  in the fresh mixture, thus  $Q$ , which is non-dimensionalized with respect to  $c_0^2$ , will be small not only because of dilution, but also due to the large sound speed in the mixture. Thus in a wide range of dilutions for helium-diluted mixtures, the low-frequency spin can be expected. Heavier diluents, especially polyatomic, would result in smaller  $\gamma$  as well as larger  $Q$ , which both correspond to high number of high-frequency unstable modes and thus would favor irregular detonation fronts. We can see in figures 3(a-b) that as  $Q$  is increased, which can be due to decrease in dilution, there are bifurcations from lower to higher frequency branches.

Now let us discuss what the effect of mixture initial pressure is on the spin detonation. One of the ways the spin detonation is observed experimentally is by lowering the initial pressure  $p_0$  of a mixture in a tube of fixed diameter and at fixed mixture composition. As the pressure is decreased a cellular detonation transforms into spin detonation after the cell size, which increases with decrease in pressure, becomes comparable to the tube diameter. The effect of the initial pressure  $p_0$  of a fresh mixture cannot be directly calculated in our model and has to be inferred from other results. The reason is that  $p_0$  is absent in the model entirely, due to the first order of the reaction rate, but since typically for realistic kinetics, the length of the reaction zone is inversely proportional to  $p_0$ , the variations of the initial pressure can be related to those of the reaction zone. For example, if  $\tilde{l}_{1/2} \sim 1/p_0$ , then the non-dimensional tube diameter  $d = \tilde{d}/\tilde{l}_{1/2} \sim \tilde{d}p_0$ . Hence lowering  $p_0$  at fixed  $\tilde{d}$  is equivalent to decreasing  $\tilde{d}$  at fixed  $p_0$  and so we can infer the role of the initial pressure from the effect of the tube diameter. In fact,  $p_0$  plays a more complex role as its variation shifts the equilibrium composition of detonation products with accompanying change in the heat effect and detonation velocity. However those dependencies are beyond the scope of the constitutive model used in this study.

## 8 Conclusions

We have investigated the instability of one-dimensional detonation wave propagating in a tube with respect to three-dimensional linear perturbations using normal-mode approach. The instability spectra are calculated exactly using high-accuracy ODE solver for the eigenfunctions and Newton-Raphson root solver for the radiation condition. The analysis shows the existence of spinning unstable modes and bifurcation from lower-frequency spin to higher-frequency spin as the mixture becomes more energetic with larger heat release  $Q$ . The behavior of the unstable modes with respect to variations of the bifurcation parameters of the problem strongly supports the possibility of the spinning instability as the origin of

the well-known spin detonation.

## 9 Acknowledgments

This work was supported by the US Air Force Office of Scientific Research (F49620-00-1-0005, program manager Dr. Arje Nachman). Additional support was provided by the US Air Force Research Laboratory Munitions Directorate, Eglin AFB (F08630-001-0002). A.R.K. is grateful to Prof. M. Short for help with code verification and for discussions about detonation stability.

## Appendix I: Definition of $\mathbf{A}_z, \mathbf{A}_r, \mathbf{A}_\theta, \mathbf{a}, \mathbf{B}_r, \mathbf{B}_\theta, \mathbf{b}, \mathbf{c}$

The following square matrices and column vectors are used to write the governing equations in shock-attached coordinates in vector form:

$$\mathbf{A}_z = \begin{bmatrix} u_z & 0 & 0 & \rho & 0 & 0 \\ 0 & u_z & 0 & 0 & 0 & 0 \\ 0 & 0 & u_z & 0 & 0 & 0 \\ 0 & 0 & 0 & u_z & 1/(\gamma\rho) & 0 \\ 0 & 0 & 0 & \gamma p & u_z & 0 \\ 0 & 0 & 0 & 0 & 0 & u_z \end{bmatrix}, \quad \mathbf{A}_r = \begin{bmatrix} u_r & \rho & 0 & 0 & 0 & 0 \\ 0 & u_r & 0 & 0 & 1/(\gamma\rho) & 0 \\ 0 & 0 & u_r & 0 & 0 & 0 \\ 0 & 0 & 0 & u_r & 0 & 0 \\ 0 & \gamma p & 0 & 0 & u_r & 0 \\ 0 & 0 & 0 & 0 & 0 & u_r \end{bmatrix}, \quad (43)$$

$$\mathbf{A}_\theta = \begin{bmatrix} u_\theta & 0 & \rho & 0 & 0 & 0 \\ 0 & u_\theta & 0 & 0 & 0 & 0 \\ 0 & 0 & u_\theta & 0 & 1/(\gamma\rho r) & 0 \\ 0 & 0 & 0 & u_\theta & 1/(\gamma\rho) & 0 \\ 0 & 0 & 0 & \gamma p & u_\theta & 0 \\ 0 & 0 & 0 & 0 & 0 & u_\theta \end{bmatrix}, \quad \mathbf{a} = \begin{bmatrix} \rho u_r \\ -u_\theta/r \\ u_r u_\theta/r \\ 0 \\ 0 \\ 0 \end{bmatrix}, \quad (44)$$

$$\mathbf{B}_r = \begin{bmatrix} u_r & \rho & 0 & 0 & 0 & 0 \\ 0 & u_r & 0 & 0 & 1/(\gamma\rho) & 0 \\ 0 & 0 & u_r & 0 & 0 & 0 \\ 0 & 0 & 0 & u_r & 0 & 0 \\ 0 & \gamma p & 0 & 0 & u_r & 0 \\ 0 & 0 & 0 & 0 & 0 & u_r \end{bmatrix}, \quad \mathbf{B}_\theta = \begin{bmatrix} u_\theta & 0 & \rho & 0 & 0 & 0 \\ 0 & u_\theta & 0 & 0 & 0 & 0 \\ 0 & 0 & u_\theta & 0 & 1/(\gamma\rho) & 0 \\ 0 & 0 & 0 & u_\theta & 0 & 0 \\ 0 & 0 & \gamma p & 0 & u_\theta & 0 \\ 0 & 0 & 0 & 0 & 0 & u_\theta \end{bmatrix}, \quad (45)$$

$$\mathbf{b} = \begin{bmatrix} 0 \\ 0 \\ 0 \\ -1 \\ 0 \\ 0 \end{bmatrix}, \quad \mathbf{c} = \begin{bmatrix} 0 \\ 0 \\ 0 \\ 0 \\ (\gamma - 1)\rho\beta\omega \\ \omega \end{bmatrix}. \quad (46)$$

## Appendix II: Radiation condition including perturbations of the reaction rate

If we separate the angular and radial dependencies of the perturbations as in (25) and use the relationship  $\hat{u}_\theta = in\hat{u}_r$ , then we get the following acoustic system in the amplitudes of perturbations depending only on  $t$  and  $z$ :

$$\frac{\partial \hat{u}_r}{\partial t} + u_b \frac{\partial \hat{u}_r}{\partial z} + \frac{\hat{p}}{\gamma \rho_b} = 0 \quad (47)$$

$$\frac{\partial \hat{u}_z}{\partial t} + u_b \frac{\partial \hat{u}_z}{\partial z} + \frac{1}{\gamma \rho_b} \frac{\partial \hat{p}}{\partial z} + \frac{\partial^2 \hat{\psi}}{\partial t^2} = 0 \quad (48)$$

$$\frac{\partial \hat{p}}{\partial t} + u_b \frac{\partial \hat{p}}{\partial z} + \gamma p_b \left( \frac{\partial \hat{u}_z}{\partial z} - k^2 \hat{u}_r \right) - (\gamma - 1) \beta \rho_b \omega_{,\lambda}^b \hat{\lambda} = 0 \quad (49)$$

Note, that  $\partial^2 \hat{\psi} / \partial t^2$  can always be absorbed into  $\hat{u}_z$  so that  $\hat{u}_z \rightarrow \hat{u}_z + \partial \hat{\psi} / \partial t$  and the equations retain their form, hence it will be dropped in the following derivations. Here  $\hat{\lambda} = \exp(\alpha t + k_z z)$  is an assumed Fourier component of the solution of the equation for  $\hat{\lambda}$ .

The above system can be written in matrix form as follows

$$\frac{\partial \mathbf{f}}{\partial t} + \mathbf{A} \frac{\partial \mathbf{f}}{\partial z} + \mathbf{B} \mathbf{f} = \mathbf{b} \hat{\lambda}, \quad (50)$$

where locally we define

$$\mathbf{f} = \begin{bmatrix} \hat{u}_r \\ \hat{u}_z \\ \hat{p} \end{bmatrix}, \quad \mathbf{A} = \begin{bmatrix} u_b & 0 & 0 \\ 0 & u_b & \frac{1}{\gamma \rho_b} \\ 0 & \gamma \rho_b c_b^2 & u_b \end{bmatrix}, \quad \mathbf{B} = \begin{bmatrix} 0 & 0 & \frac{1}{\gamma \rho_b} \\ 0 & 0 & 0 \\ -\gamma \rho_b k^2 c_b^2 & 0 & 0 \end{bmatrix}, \quad \mathbf{b} = \begin{bmatrix} 0 \\ 0 \\ a_0 \end{bmatrix}, \quad (51)$$

and  $a_0 = (\gamma - 1) \beta \rho_b \omega_{,\lambda}^b$ ,  $c_b^2 = p_b / \rho_b$ ,  $\alpha = \omega_{,\lambda}^b - k_z u_b$ . The system (50) can be diagonalized if written in terms of the Riemann invariants  $r_i = \mathbf{l}_i \cdot \mathbf{f}$ , i.e. the dot products of the left eigenvectors of the matrix  $\mathbf{A}$  and the vector  $\mathbf{f}$ . The eigenvectors are

$$\mathbf{l}_1 = [0, \gamma \rho_b c_b, 1], \quad \mathbf{l}_2 = [1, 0, 0], \quad \mathbf{l}_3 = [0, -\gamma \rho_b c_b, 1]. \quad (52)$$

Then we get  $\mathbf{l}_i \mathbf{A} = m_i \mathbf{l}_i$  with

$$m_1 = u_b + c_b, \quad m_2 = u_b, \quad m_3 = u_b - c_b, \quad (53)$$

and the diagonalized system in  $r_i$  becomes

$$\frac{\partial r_i}{\partial t} + m_i \frac{\partial r_i}{\partial z} + b_{1i} = b_{2i} \hat{\lambda}, \quad \text{for } i = 1, 2, 3, \quad (54)$$

where

$$\mathbf{r} = \begin{bmatrix} \hat{p} + \gamma \rho_b c_b \hat{u}_z \\ \hat{u}_r \\ \hat{p} - \gamma \rho_b c_b \hat{u}_z \end{bmatrix}, \quad \mathbf{b}_1 = \mathbf{l} \mathbf{B} \mathbf{f} = \begin{bmatrix} -\gamma \rho_b c_b^2 k^2 \hat{u}_r \\ \frac{\hat{p}}{\gamma \rho_b} \\ -\gamma \rho_b c_b^2 k^2 \hat{u}_r \end{bmatrix}, \quad \mathbf{b}_2 = \mathbf{l} \mathbf{b} = \begin{bmatrix} a_0 \\ 0 \\ a_0 \end{bmatrix} \quad (55)$$

If we denote  $L_+ = \partial/\partial t + (u_b + c_b) \partial/\partial z$ ,  $L_- = \partial/\partial t + (u_b - c_b) \partial/\partial z$ , and  $L_0 = \partial/\partial t + u_b \partial/\partial z$ , the equations for  $r_i$  can be written in expanded form as

$$L_+(r_1) - \gamma \rho_b c_b^2 k^2 r_2 = a_0 \hat{\lambda}, \quad (56)$$

$$L_0(r_2) + \frac{1}{2\gamma \rho_b} (r_1 + r_3) = 0, \quad (57)$$

$$L_-(r_3) - \gamma \rho_b c_b^2 k^2 r_2 = a_0 \hat{\lambda}. \quad (58)$$

If we take  $L_-$  of (56),  $L_+$  of (58), add them, and make use of (57), we get a single equation in  $r_1 + r_3 = 2\hat{p}$ , which is the non-homogeneous Klein-Gordon equation

$$\frac{\partial^2 \hat{p}}{\partial t^2} + 2u_b \frac{\partial^2 \hat{p}}{\partial t \partial z} - (c_b^2 - u_b^2) \frac{\partial^2 \hat{p}}{\partial z^2} + (c_b k)^2 \hat{p} - a_0 \omega_{,\lambda}^b e^{\alpha t + k_z z} = 0 \quad (59)$$

The solution of this equation is

$$\hat{p} = A_1 e^{\alpha t + s z} + A_0 e^{\alpha t + k_z z}, \quad (60)$$

where  $A_1$  is an unknown constant,  $A_0 = a_0 \omega_{,\lambda}^b / ((\omega_{,\lambda}^b)^2 + (k^2 - k_z^2) c_\infty^2)$ . The complex wavenumber  $s$  is given by equation (37) and satisfies the boundedness condition as discussed in the main text above.

If we now add equations (56) and (58) and write  $L_+$  and  $L_-$  explicitly, we get the following equation

$$\frac{\partial \hat{p}}{\partial t} + u_b \frac{\partial \hat{p}}{\partial z} + \gamma \rho_b c_b^2 \frac{\partial \hat{u}_z}{\partial z} - \gamma \rho_b c_b^2 k^2 \hat{u}_r = a_0 \hat{\lambda}. \quad (61)$$

We separate the exponential time dependence as  $\hat{p} = p' \exp(\alpha t)$  and so on, to get

$$\alpha p' + u_b \frac{dp'}{dz} + \gamma \rho_b c_b^2 \frac{du'_z}{dz} - \gamma \rho_b c_b^2 k^2 u'_r = a_0 \lambda'. \quad (62)$$

Differentiating (60) we get

$$\frac{dp'}{dz} = s(p' - A_0\lambda') + A_0k_z\lambda'. \quad (63)$$

Inserting this expression into (62) we obtain the general radiation condition (for  $\lambda' \neq 0$ )

$$\frac{du'_z}{dz} - k^2u'_r + \frac{\alpha + su_b}{\gamma\rho_b c_b^2}p' - \mu\lambda' = 0, \quad (64)$$

where

$$\mu = \frac{\gamma - 1}{\gamma} \frac{\beta\omega_{,\lambda}^b}{c_b^2} \left[ 1 + \frac{\omega_{,\lambda}^b(s - k_z)}{(\omega_{,\lambda}^b)^2 + (k^2 - k_z^2)c_b^2} \right], \quad (65)$$

$k_z = (\omega_{,\lambda}^b - \alpha)/u_b$ , and  $s = \left( \alpha u_b + c_b [\alpha^2 + k^2(c_b^2 - u_b^2)]^{1/2} \right) / (c_b^2 - u_b^2)$

In previous studies (except in Lee & Stewart [14]) the term proportional to  $\lambda'$  has been dropped on the grounds of its presumed exponential smallness in the burnt region. It is clear from (65) that  $\mu = O(\omega_{,\lambda}^b) = O(\exp(-\theta/c_b^2))$ , provided that other factors, including the square bracket are  $O(1)$ . Thus the assumption is justified at large  $\theta$ , but there has been no rigorous *a priori* justification for neglecting the term at small or moderate  $\theta$ . A possible argument in favor of neglecting the term is that as  $z \rightarrow -\infty$ ,  $\lambda' \sim \exp(k_z z) \sim \exp(\text{Re}(k_z)z)$  and  $\text{Re}(k_z) = -(\exp(-\theta/c_b^2) + \alpha_r)/u_b$ , therefore  $\lambda'$  decays exponentially with both  $\theta$  and  $z$ :

$$\mu\lambda' \sim \exp \left[ -\frac{\theta}{c_b^2} - \frac{z}{u_b} \left( \alpha_r + \exp \left( -\frac{\theta}{c_b^2} \right) \right) \right].$$

When  $\theta$  is large the decay of  $\mu\lambda'$  is due to  $\mu$ , while at small  $\theta$  it is due to  $\lambda'$ . The latter is exactly what is observed in numerical calculations, which show that variations in  $\lambda'$  are typically concentrated to a narrow region near the shock. Still, these estimates are good only if all other terms in the radiation condition are much larger than  $\mu\lambda'$ . To find out if the latter is true, one would have to find the asymptotics of the entire problem.

Reduction of the radiation condition (64) to the one-dimensional case can be done through the integration of (64) over  $z$  at  $k = 0$  and shows that it is exactly the same condition as given in Lee & Stewart (1990). One should only keep in mind that  $\int (du'_z/dz)dz = u'_z + \alpha$  which is due to the difference in definitions of  $u_z$ .

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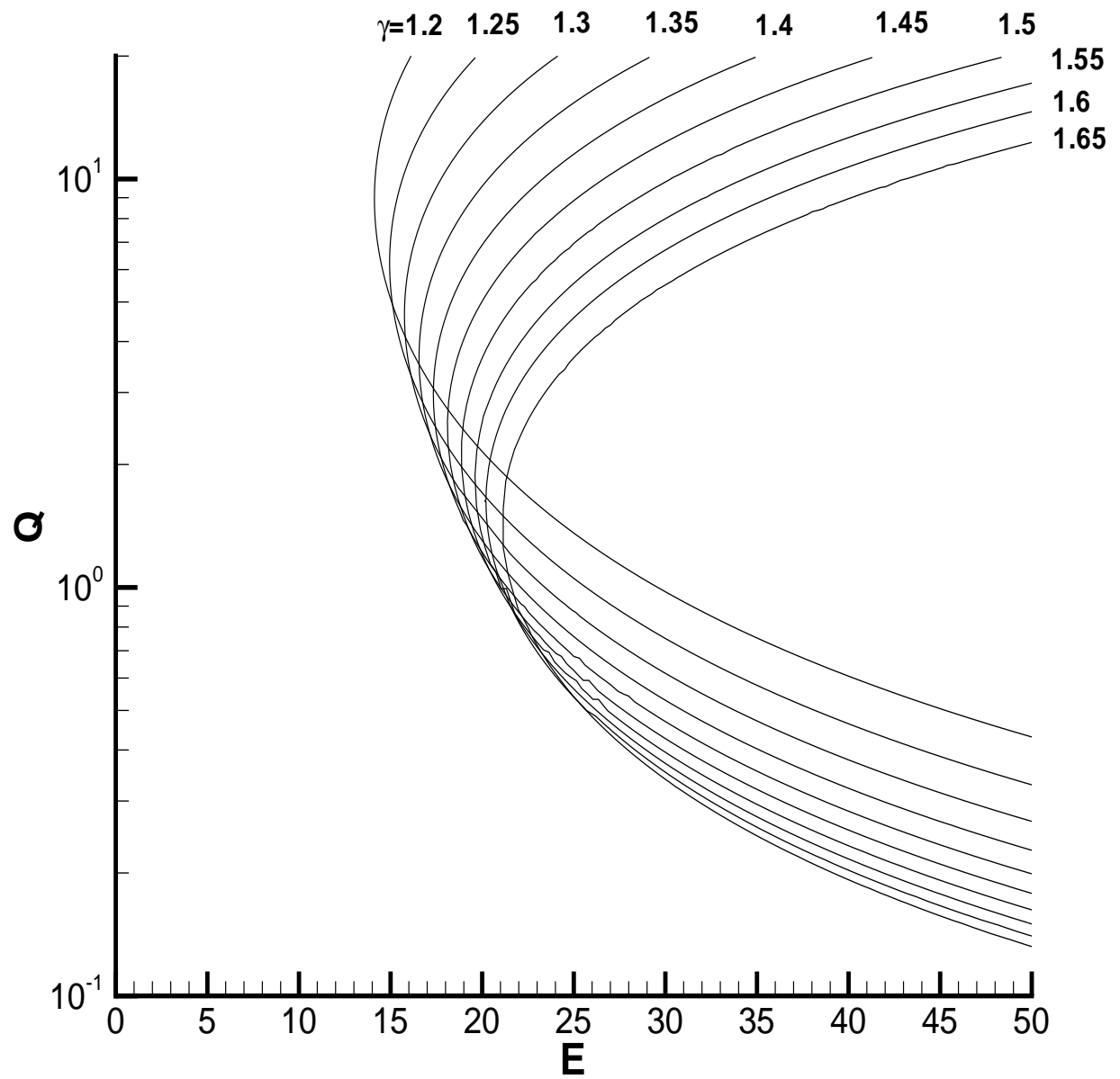


Figure 2: The effect of the adiabatic exponent  $\gamma$  on the one-dimensional neutral stability boundary.

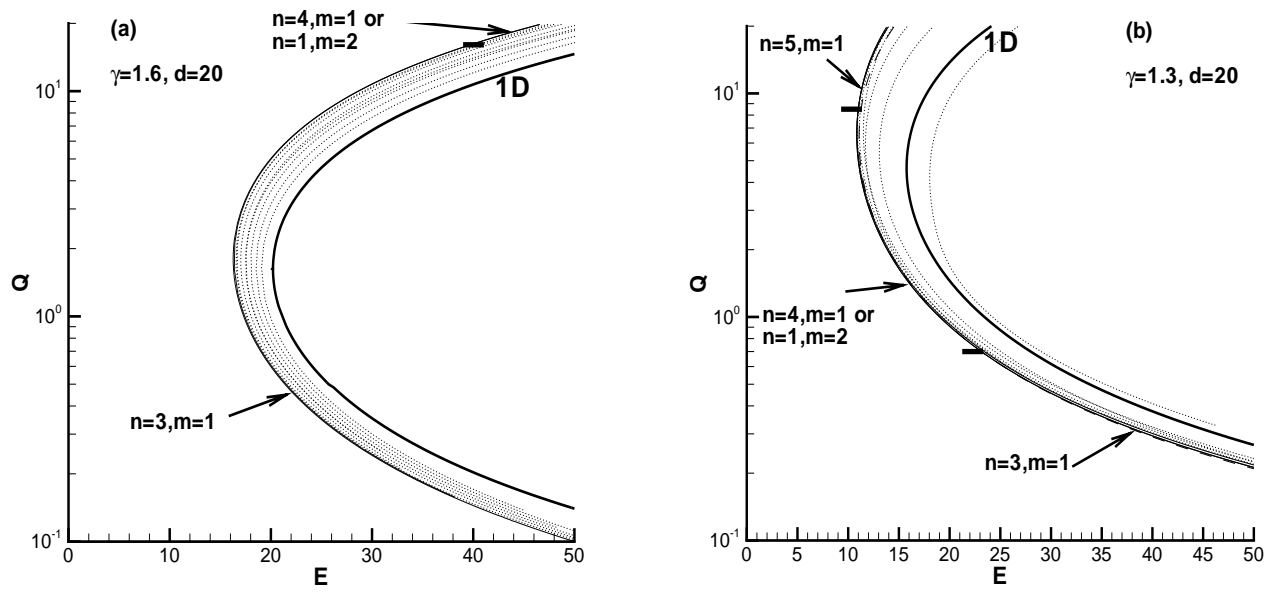


Figure 3: (a) - neutral stability boundaries in  $QE$ -plane for spinning modes in a tube with diameter  $d = 20$  at  $\gamma = 1.6$ ; (b) - same for  $\gamma = 1.3$ .

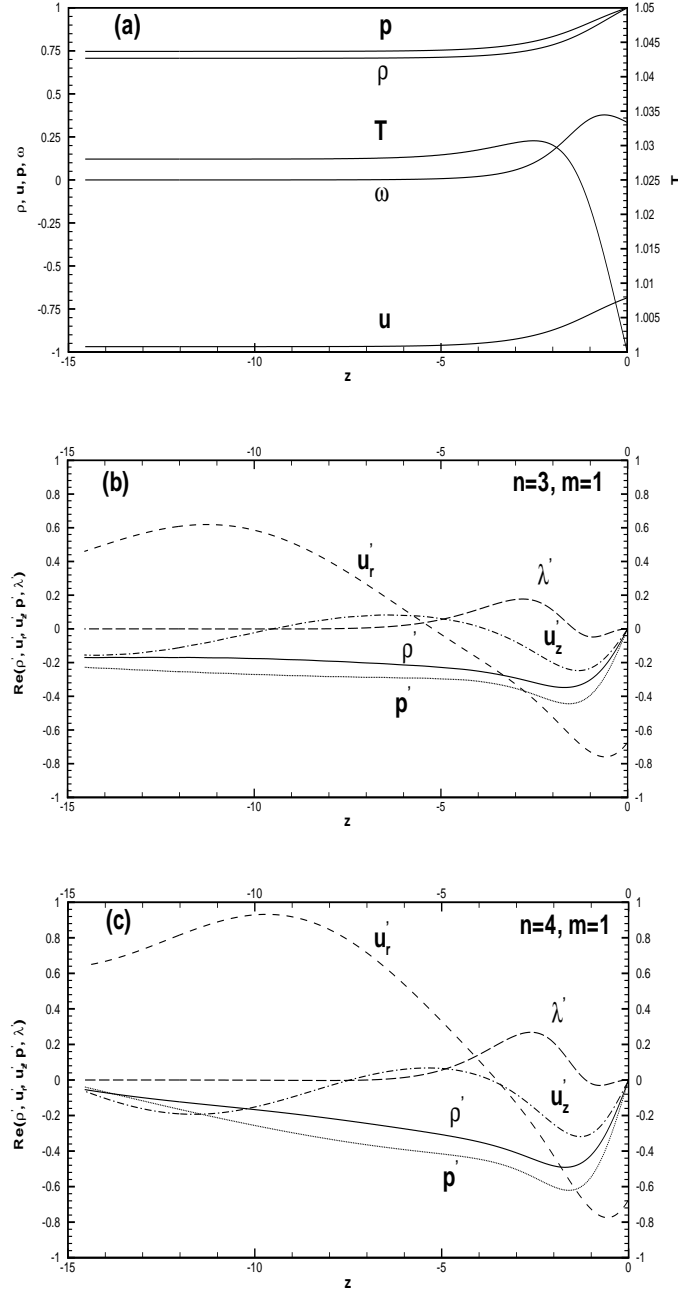


Figure 4: Steady state profiles (a) and real parts of eigenfunctions, (b) and (c), at  $Q = 0.69$ ,  $E = 23.23$ ,  $\gamma = 1.3$ , and  $d = 20$  (corresponds to bifurcation point on figure 3(b)). On figures (b) and (c) solid line is  $Re(\rho')$ , dashed  $Re(u_r')$ , dashdot  $Re(u_z')$ , dotted  $Re(p')$ , and long dash  $Re(\lambda')$ .

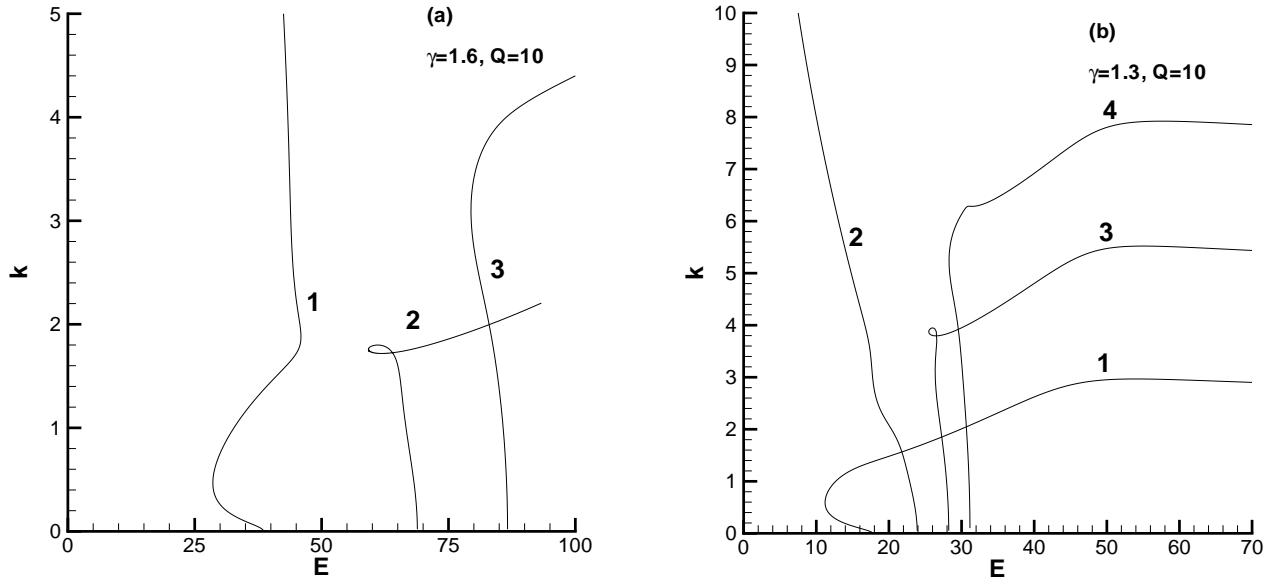


Figure 5:  $Ek$  neutral stability curves for the first few temporal roots at  $Q = 10$  and  $\gamma = 1.6$  (a) and  $\gamma = 1.3$  (b).

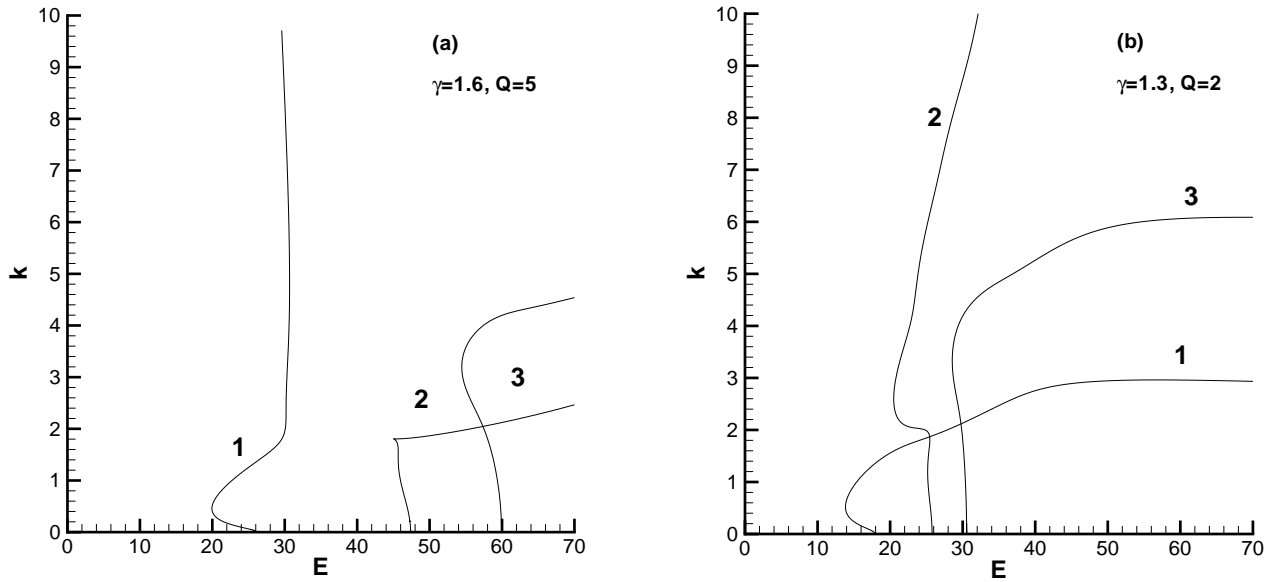


Figure 6:  $Ek$  neutral stability curves for the first several roots at  $Q = 5$ ,  $\gamma = 1.6$  (a), and  $Q = 2$ ,  $\gamma = 1.3$  (b).

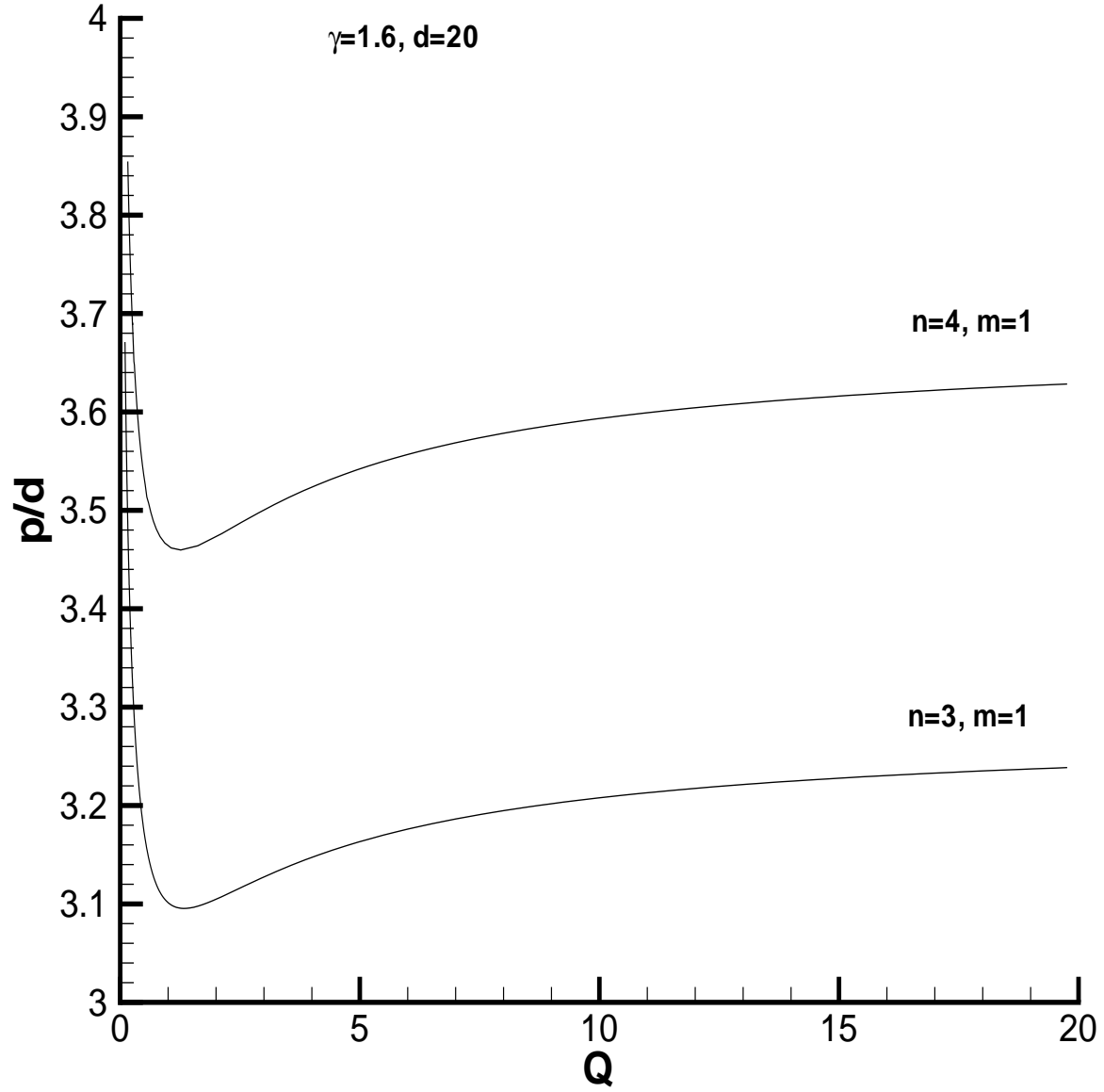


Figure 7: Pitch ratio  $p/d$  vs the heat release  $Q$  along the neutral stability boundaries for  $n = 3, m = 1$  and  $n = 4, m = 1$  corresponding to figure 3(a).

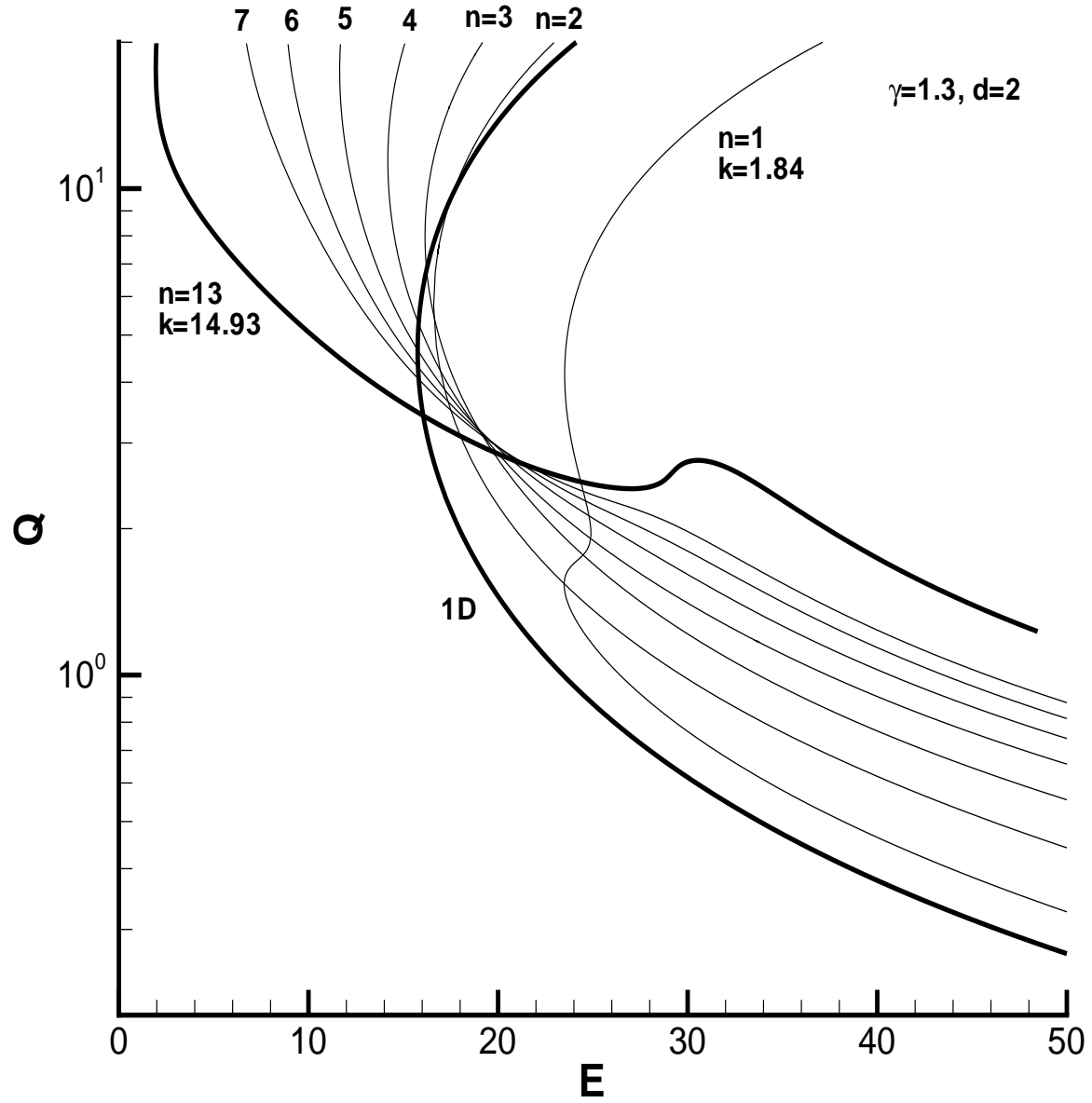


Figure 8: Stability behaviour of detonation in a small tube with diameter  $d = 2$ : QE neutral stability curves for modes  $n = 1$  through  $n = 13$  at  $m = 1$  and  $\gamma = 1.3$ .