Effects of Diffusion on the Dynamics of Detonation

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*and contributions from many others

Motivation

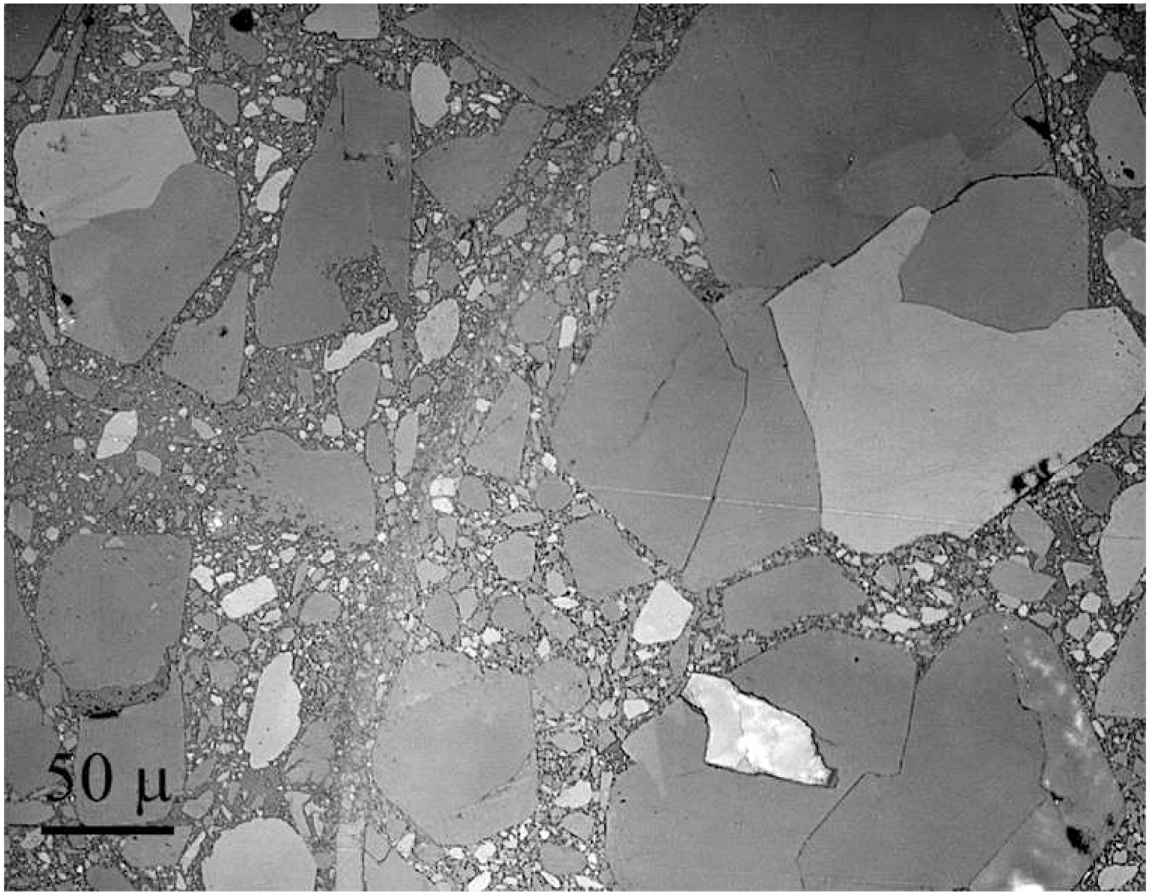
- Computational tools are critical in modeling of high speed reactive flow.
- Steady wave calculations reveal sub-micron scale structures in detonations with detailed kinetics (Powers and Paolucci, *AIAA J.*, 2005).
- Small structures are continuum manifestation of molecular collisions.
- We explore the transient behavior of detonations with *fully resolved* detailed kinetics.

Verification and Validation

- *verification:* solving the equations right (math).
- *validation:* solving the right equations (physics).
- Main focus here on verification
- Some limited validation possible, but detailed validation awaits more robust measurement techniques.
- Verification and validation always necessary but never sufficient: finite uncertainty must be tolerated.

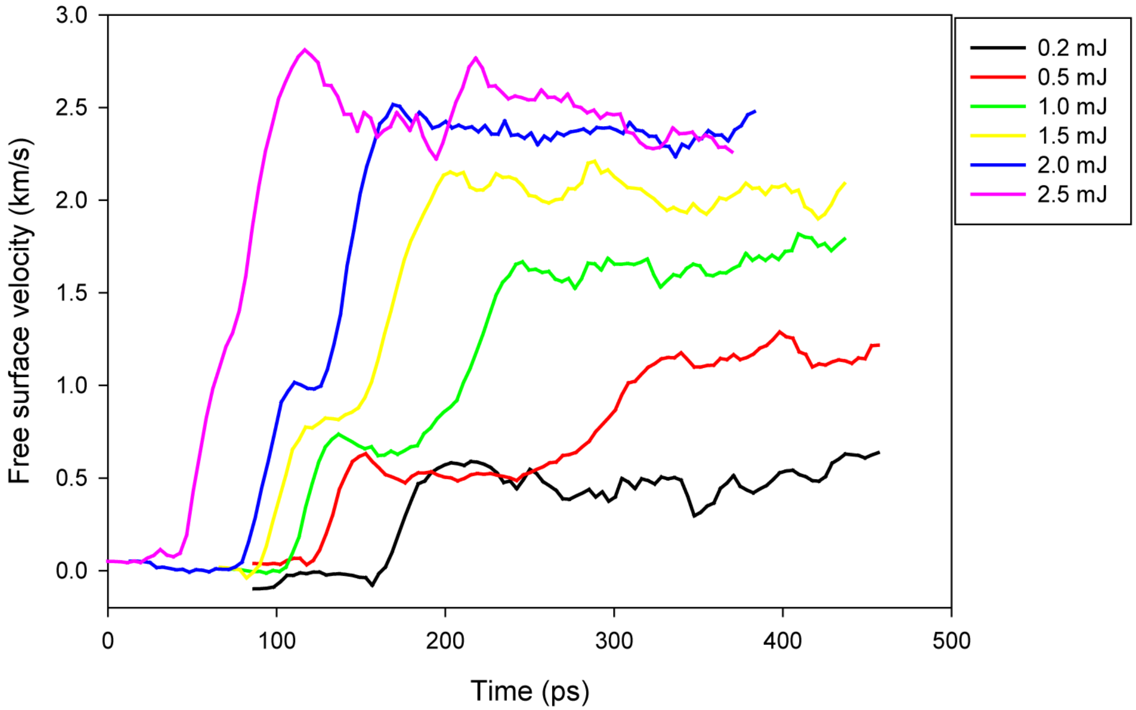
Some Length Scales inherent in PBXs

Micrograph of PBX 9501 (from C. Skidmore)



Some Length Scales Due to Diffusion

Shock Rise in Aluminum (from V. Whitley)



$10\ ps$ rise time at $10\ km/s$ yields scale of $10^{-7}\ m$.

Modeling Issues for PBXs:

- Inherently 3D, multi-component mixture,
- Massive disparity in scales,
- Many parameters are needed and many are unknown*:
  - elastic constants
  - equation of states
  - thermal conductivities, viscosities for constituents
  - heat capacities
  - reaction rates
  - species diffusion

*see Menikoff and Sewell, CTM 2002

Before climbing Everest, we need to step back a bit...

Let’s examine detonation dynamics of gases...

1. Inviscid, one-step Arrhenius chemistry
2. Inviscid, detailed chemistry
3. Diffusive, one-step Arrhenius chemistry
4. Diffusive, detailed chemistry

Model: Reactive Euler Equations

- one-dimensional,
- unsteady,
- inviscid,
- detailed mass action kinetics with Arrhenius temperature dependency,
- ideal mixture of calorically imperfect ideal gases

General Review of Pulsating Detonations

- Erpenbeck, *Phys. Fluids*, 1962,
- Fickett and Wood, *Phys. Fluids*, 1966,
- Lee and Stewart, *JFM*, 1990,
- Bourlioux, *et al.*, *SIAM J. Appl. Math.*, 1991,
- He and Lee, *Phys. Fluids*, 1995,
- Short, *SIAM J. Appl. Math.*, 1997,
- Sharpe, *Proc. R. Soc.*, 1997.

Review of Recent Work of Special Relevance

- Kasimov and Stewart, *Phys. Fluids*, 2004: published detailed discussion of limit cycle behavior with shock-fitting; error $\sim O(\Delta x)$.
- Ng, Higgins, Kiyanda, Radulescu, Lee, Bates, and Nikiforakis, *CTM*, in press, 2005: in addition, considered transition to chaos; error $\sim O(\Delta x)$.
- Present study similar to above, but error $\sim O(\Delta x^5)$.

Model: Reactive Euler Equations

- one-dimensional,
- unsteady,
- inviscid,
- one step kinetics with finite activation energy,
- calorically perfect ideal gases with identical molecular masses and specific heats.

Model: Reactive Euler Equations

$$
\begin{aligned}
\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial \xi}(\rho u) &= 0, \\
\frac{\partial}{\partial t}(\rho u) + \frac{\partial}{\partial \xi}\left(\rho u^2 + p\right) &= 0, \\
\frac{\partial}{\partial t}\left(\rho\left(e + \frac{1}{2}u^2\right)\right) + \frac{\partial}{\partial \xi}\left(\rho u\left(e + \frac{1}{2}u^2 + \frac{p}{\rho}\right)\right) &= 0, \\
\frac{\partial}{\partial t}(\rho\lambda) + \frac{\partial}{\partial \xi}(\rho u\lambda) &= \alpha\rho(1-\lambda)\exp\left(-\frac{\rho E}{p}\right), \\
e &= \frac{1}{\gamma - 1}\frac{p}{\rho} - \lambda q.
\end{aligned}
$$

Unsteady Shock Jump Equations

$$
\begin{aligned}
\rho_s(D(t) - u_s) &= \rho_o(D(t) - u_o), \\
p_s - p_o &= (\rho_o(D(t) - u_o))^2 \left(\frac{1}{\rho_o} - \frac{1}{\rho_s} \right), \\
e_s - e_o &= \frac{1}{2}(p_s + p_o) \left(\frac{1}{\rho_o} - \frac{1}{\rho_s} \right), \\
\lambda_s &= \lambda_o.
\end{aligned}
$$

Model Refinement

- Transform to shock attached frame via

$$x = \xi - \int_0^t D(\tau)d\tau,$$

- Use jump conditions to develop shock-change equation for shock acceleration:

$$\frac{dD}{dt} = -\left(\frac{d(\rho_s u_s)}{dD}\right)^{-1} \left(\frac{\partial}{\partial x}\left(\rho u(u - D) + p\right)\right).$$

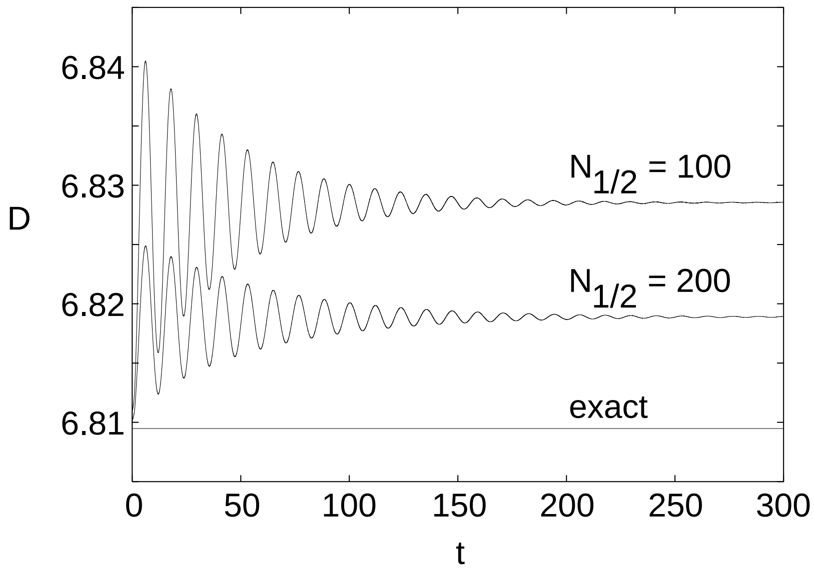
Numerical Method

- point-wise method of lines,
- uniform spatial grid,
- fifth order spatial discretization (WENO5M) takes PDEs into ODEs in time only,
- fifth order explicit Runge-Kutta temporal discretization to solve ODEs.
- details in Henrick, Aslam, Powers, *JCP*, 2006.

Numerical Simulations

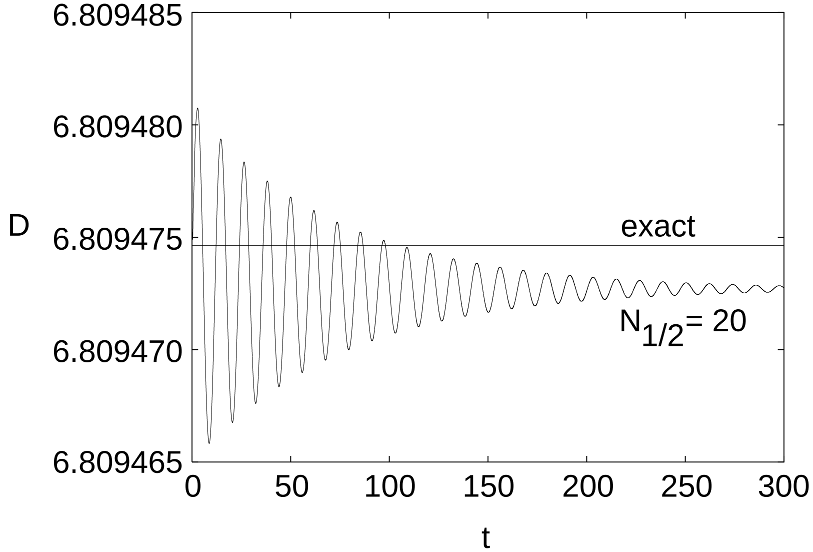
- $\rho_o = 1, p_o = 1, L_{1/2} = 1, q = 50, \gamma = 1.2,$
- Activation energy, E, a variable bifurcation parameter, $25 \le E \le 28.4,$
- CJ velocity: $D_{CJ} = \sqrt{11} + \sqrt{\frac{61}{5}} \approx 6.80947463,$
- from 10 to 200 points in $L_{1/2}$,
- initial steady CJ state perturbed by truncation error,
- integrated in time until limit cycle behavior realized.

Stable Case, $E = 25$: Kasimov's Shock-Fitting



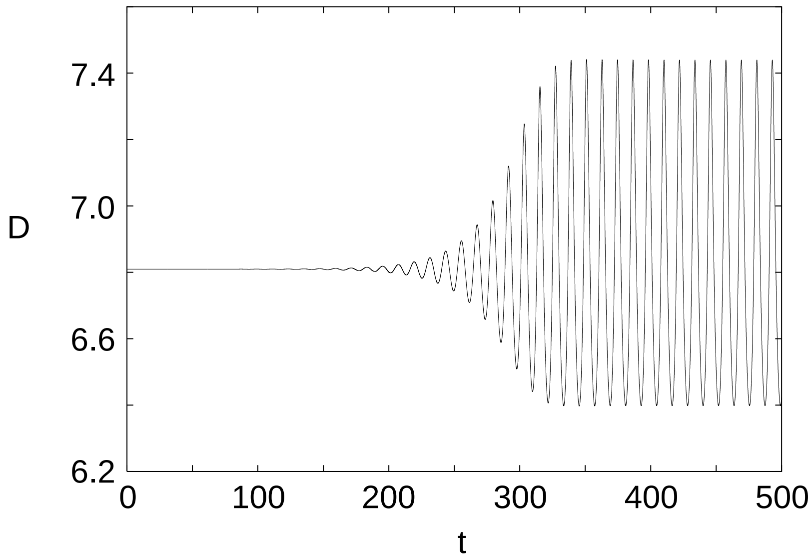
- $N_{1/2} = 100,\ 200,$
- minimum error in D: $\sim 9.40 \times 10^{-3}$,
- Error in D converges at $O(\Delta x^{1.01})$.

Stable Case, $E = 25$: Improved Shock-Fitting



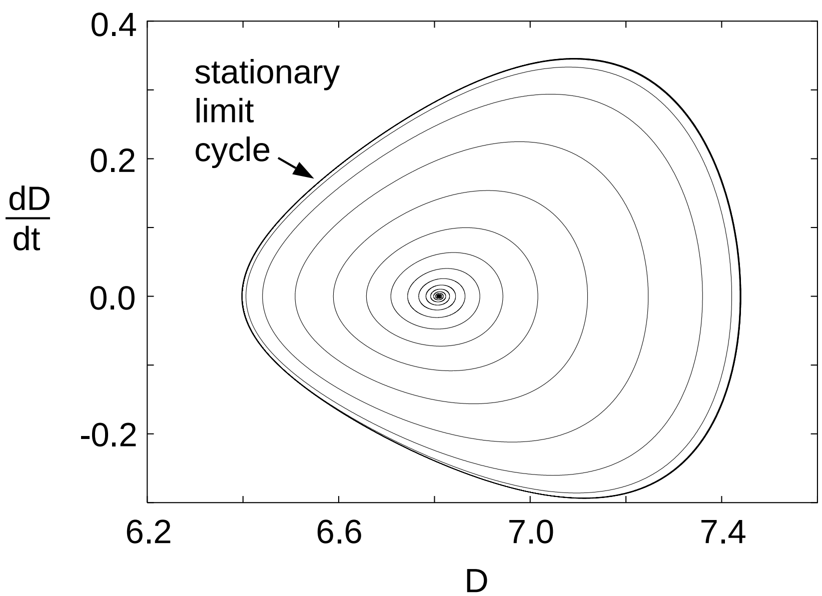
- $N_{1/2} = 20,\ 40,$
- minimum error in D: $\sim\ 6.00 \times 10^{-8}$, for $N_{1/2} = 40.$
- Error in D converges at $O(\Delta x^{5.01})$.

Linearly Unstable, Non-linearly Stable Case: $E = 26$



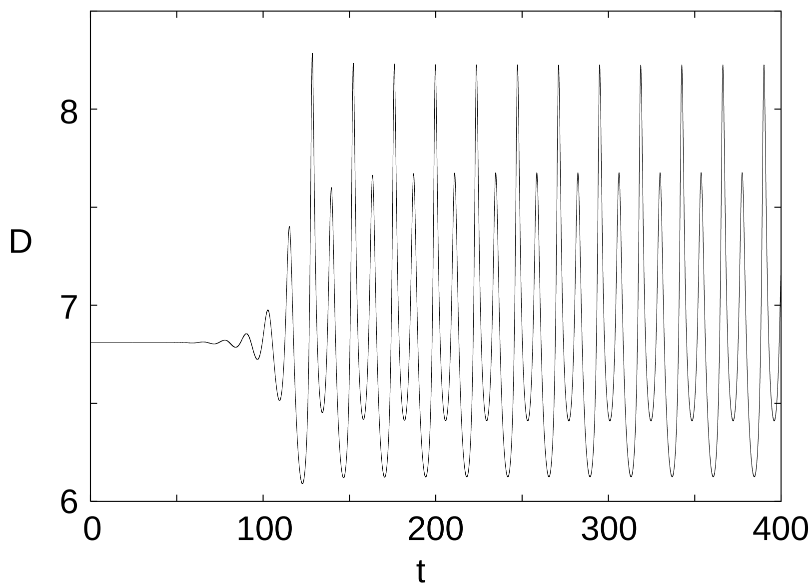
- One linearly unstable mode, stabilized by non-linear effects,
- Growth rate and frequency match linear theory to five decimal places.

$D, \frac{dD}{dt}$ **Phase Plane:** $E = 26$



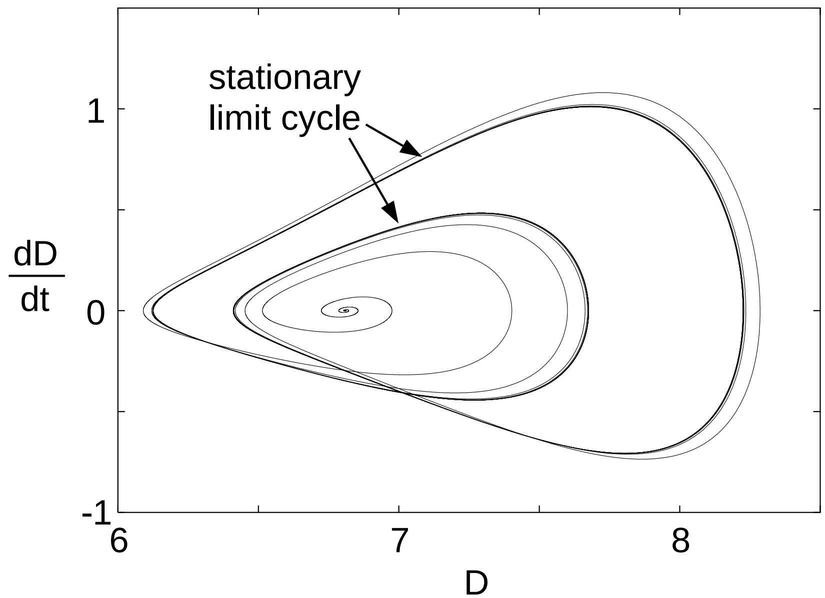
- Unstable spiral at early time, stable period-1 limit cycle at late time,
- Bifurcation point of $E = 25.265 \pm 0.005$ agrees with linear stability theory.

Period Doubling: $E = 27.35$



- $N_{1/2} = 20$,
- Bifurcation to period-2 oscillation at $E = 27.1875 \pm 0.0025$.

$D, \ \frac{dD}{dt}$ Phase Plane: $E = 27.35$



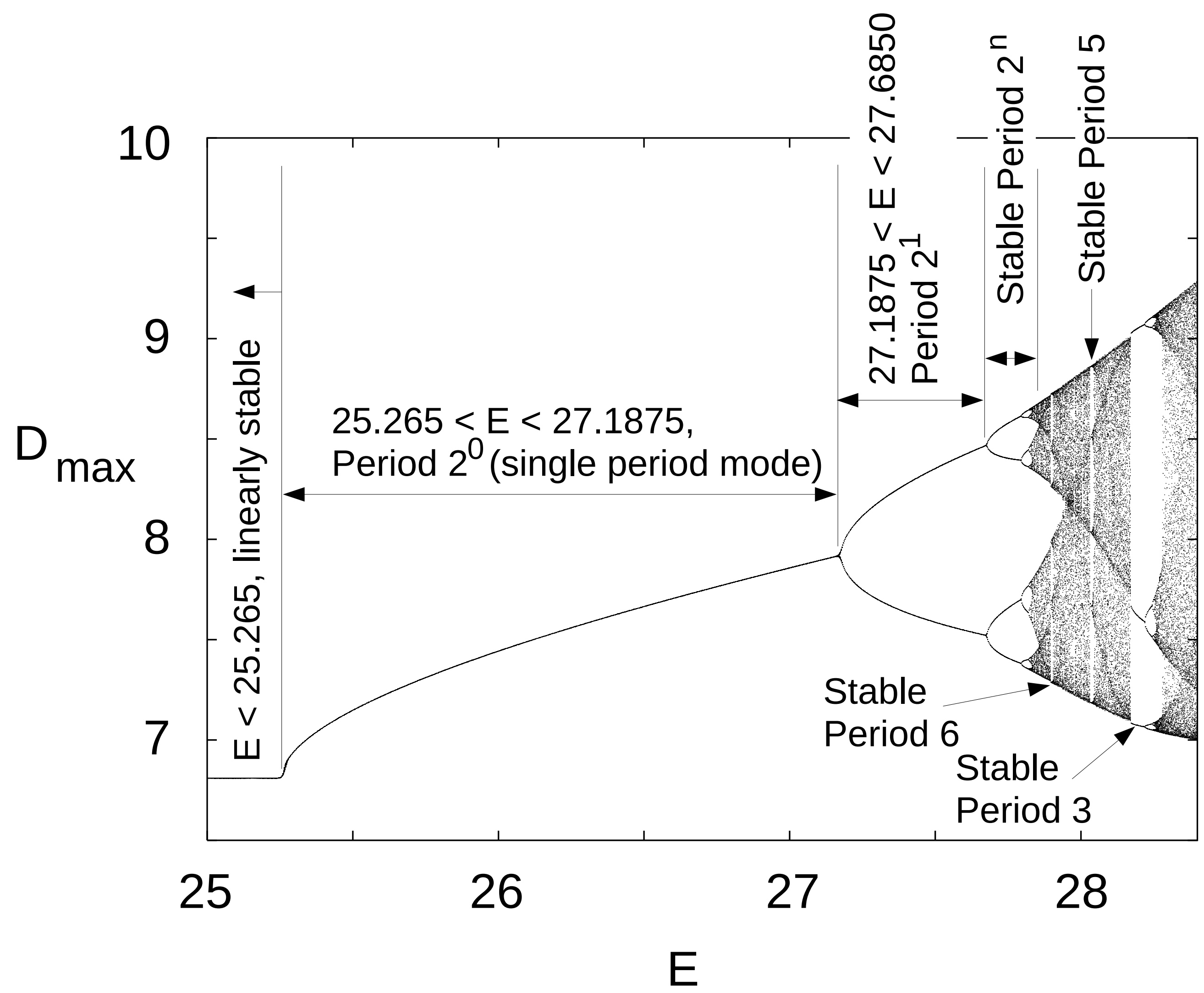
- Long time period-2 limit cycle,
- Similar to independent results of Sharpe and Ng.

Transition to Chaos and Feigenbaum's Number

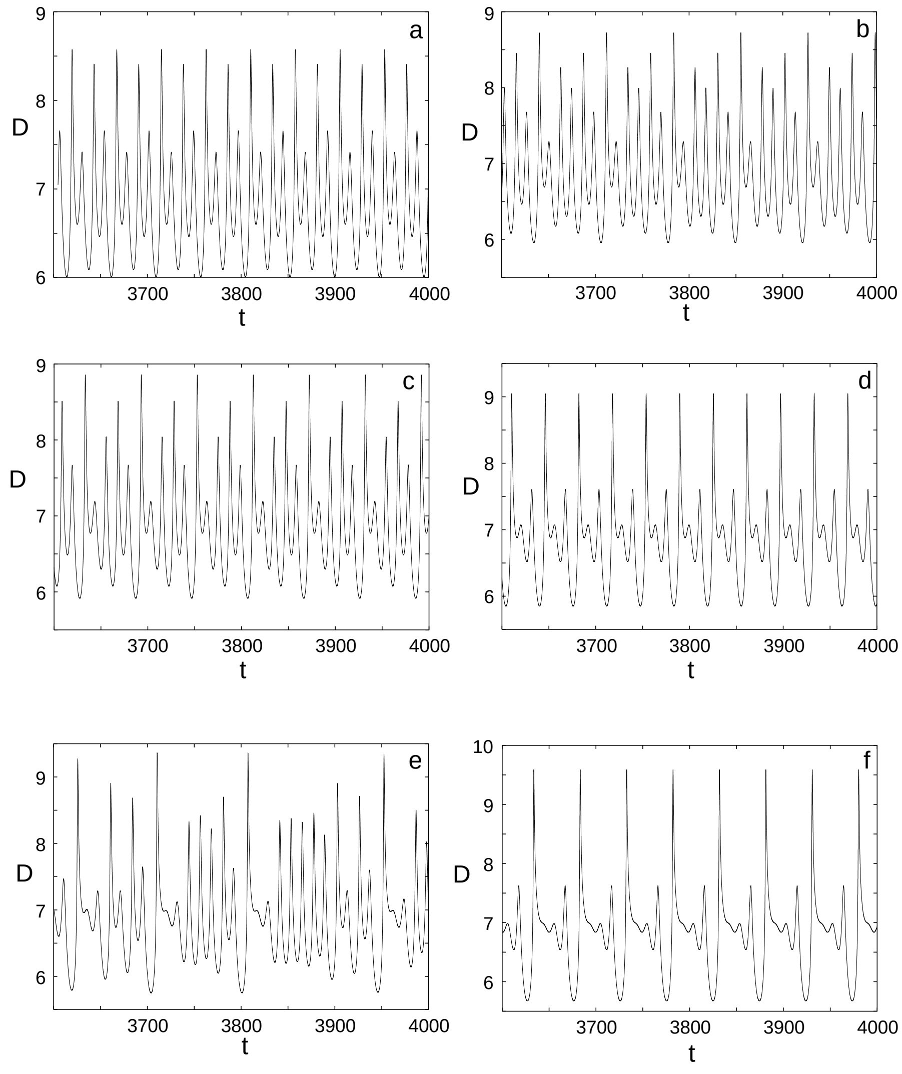
$$\lim_{n\to\infty} \delta_n = \frac{E_n - E_{n-1}}{E_{n+1} - E_n} = 4.669201\ldots.$$

n	E_n	$E_{n+1} - E_i$	δ_n
0	25.265 ± 0.005	-	-
1	27.1875 ± 0.0025	1.9225 ± 0.0075	3.86 ± 0.05
2	27.6850 ± 0.001	0.4975 ± 0.0325	4.26 ± 0.08
3	27.8017 ± 0.0002	0.1167 ± 0.0012	4.66 ± 0.09
4	27.82675 ± 0.00005	0.02505 ± 0.00025	-
$\vdots$	$\vdots$	$\vdots$	$\vdots$
∞			$4.669201\ldots$

Bifurcation Diagram



D versus t for Increasing E



Model: Reactive Euler PDEs with Detailed Kinetics

$$
\begin{aligned}
\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x}(\rho u) &= 0, \\
\frac{\partial}{\partial t}(\rho u) + \frac{\partial}{\partial x}\left(\rho u^2 + p\right) &= 0, \\
\frac{\partial}{\partial t}\left(\rho\left(e + \frac{u^2}{2}\right)\right) + \frac{\partial}{\partial x}\left(\rho u\left(e + \frac{u^2}{2} + \frac{p}{\rho}\right)\right) &= 0, \\
\frac{\partial}{\partial t}(\rho Y_i) + \frac{\partial}{\partial x}(\rho u Y_i) &= M_i \dot{\omega}_i, \\
p &= \rho \Re T \sum_{i=1}^{N} \frac{Y_i}{M_i}, \\
e &= e(T, Y_i), \\
\dot{\omega}_i &= \dot{\omega}_i(T, Y_i).
\end{aligned}
$$

Computational Methods

- Steady wave structure
  - LSODE solver with IMSL DNEQNF for root finding
  - Ten second run time on single processor machine.
  - see Powers and Paolucci, *AIAA J.*, 2005.
- Unsteady wave structure
  - Shock fitting coupled with a high order method for continuous regions
  - see Henrick, Aslam, Powers, *J. Comp. Phys.*, 2006, for full details on shock fitting

Ozone Reaction Kinetics

Reaction	a_j^f, a_j^r	β_j^f, β_j^r	E_j^f, E_j^r
$O_3 + M \leftrightarrows O_2 + O + M$	6.76×10^6	2.50	1.01×10^{12}
	1.18×10^2	3.50	0.00
$O + O_3 \leftrightarrows 2O_2$	4.58×10^6	2.50	2.51×10^{11}
	1.18×10^6	2.50	4.15×10^{12}
$O_2 + M \leftrightarrows 2O + M$	5.71×10^6	2.50	4.91×10^{12}
	2.47×10^2	3.50	0.00

see Margolis, *J. Comp. Phys.*, 1978, or Hirschfelder, *et al.*, *J. Chem. Phys.*, 1953.

Validation: Comparison with Observation

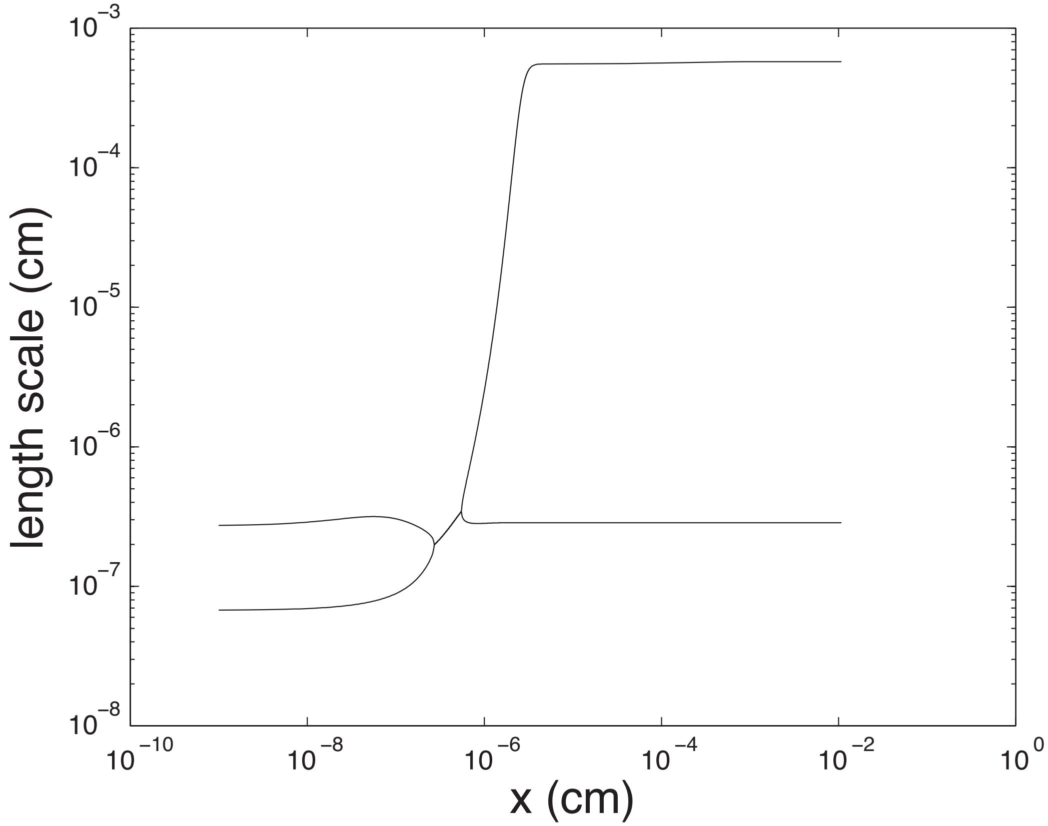
- Streng, *et al.*, *J. Chem. Phys.*, 1958.
- $p_o = 1.01325 \times 10^6\ dyne/cm^2$, $T_o = 298.15\ K$, $Y_{O_3} = 1, Y_{O_2} = 0, Y_O = 0$.

Value	Streng, *et al.*	this study
D_{CJ}	$1.863 \times 10^5\ cm/s$	$1.936555 \times 10^5\ cm/s$
T_{CJ}	$3340\ K$	$3571.4\ K$
p_{CJ}	$3.1188 \times 10^7\ dyne/cm^2$	$3.4111 \times 10^7\ dyne/cm^2$

Slight overdrive to preclude interior sonic points.

Stable Strongly Overdriven Case: Length Scales

$D = 2.5 \times 10^5 \; cm/s.$



Mean-Free-Path Estimate

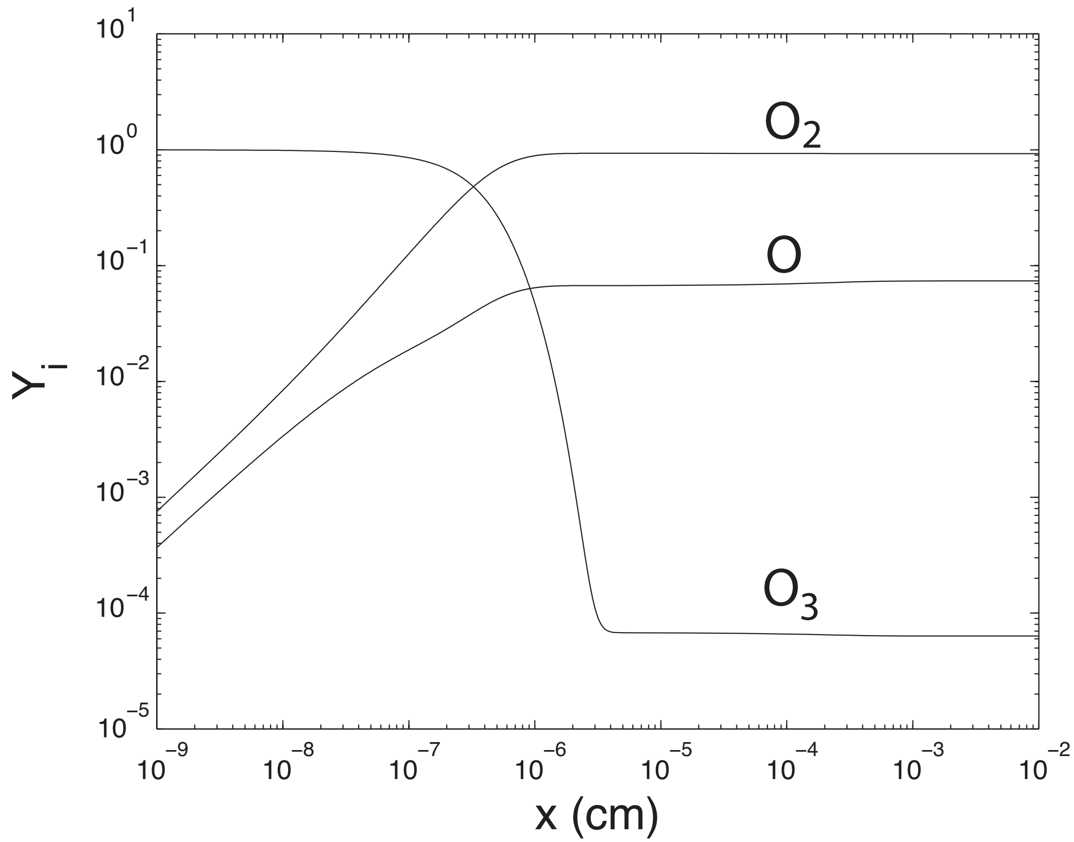
- The mixture mean-free-path scale is the cutoff *minimum* length scale associated with continuum theories.

- A simple estimate for this scale is given by *Vincenti and Kruger, '65*:

$$\ell_{mfp} = \frac{M}{\sqrt{2}\mathcal{N}\pi d^2 \rho} \sim 10^{-7}\, cm.$$

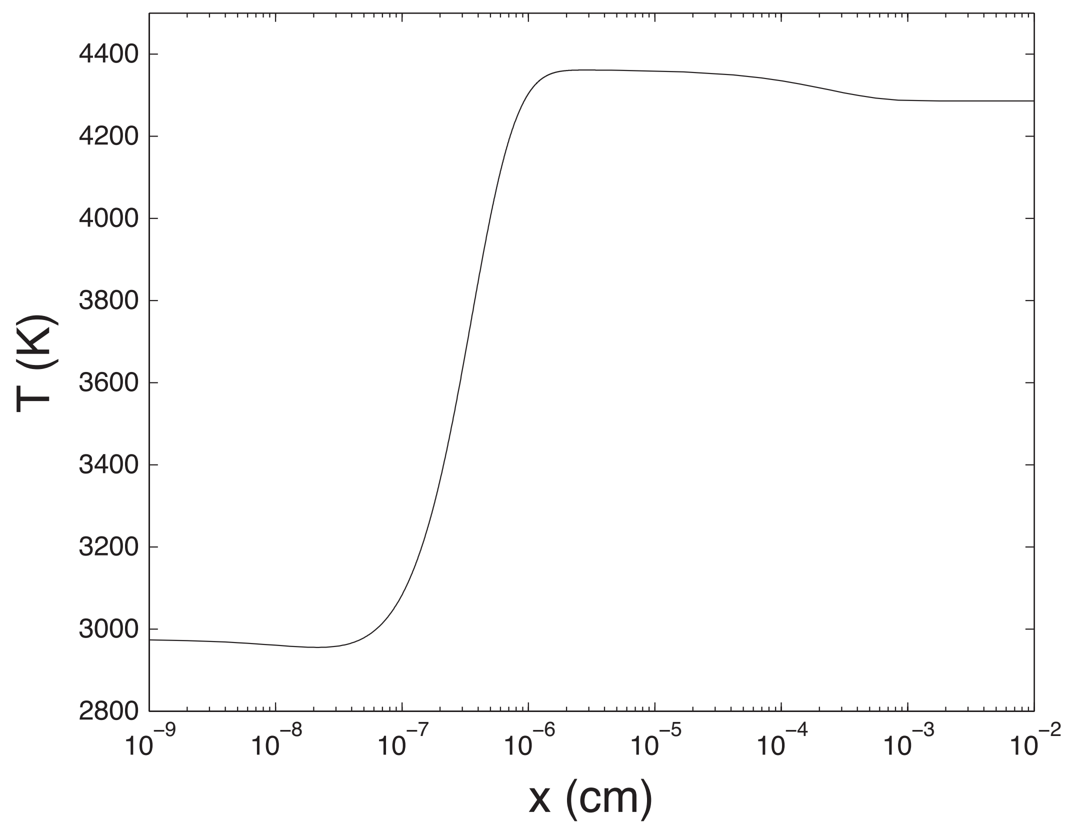
Stable Strongly Overdriven Case: Mass Fractions

$$D = 2.5 \times 10^5 \ cm/s.$$



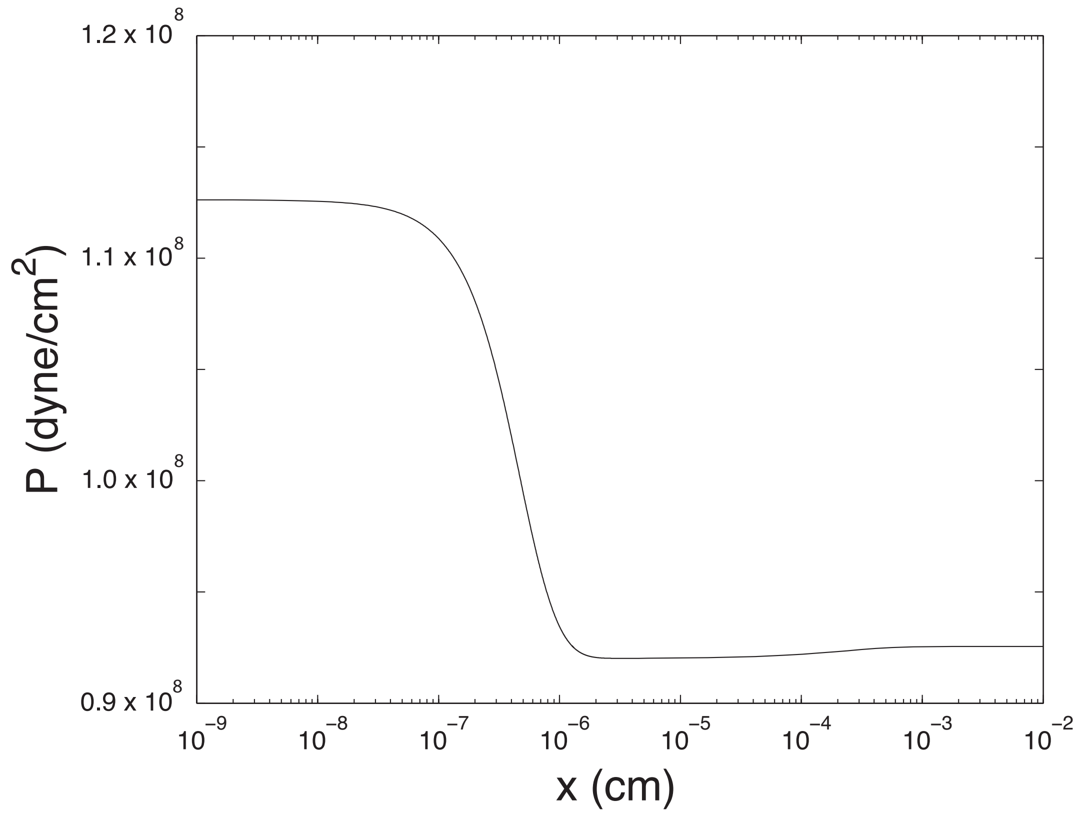
Stable Strongly Overdriven Case: Temperature

$$D = 2.5 \times 10^5 \ cm/s.$$



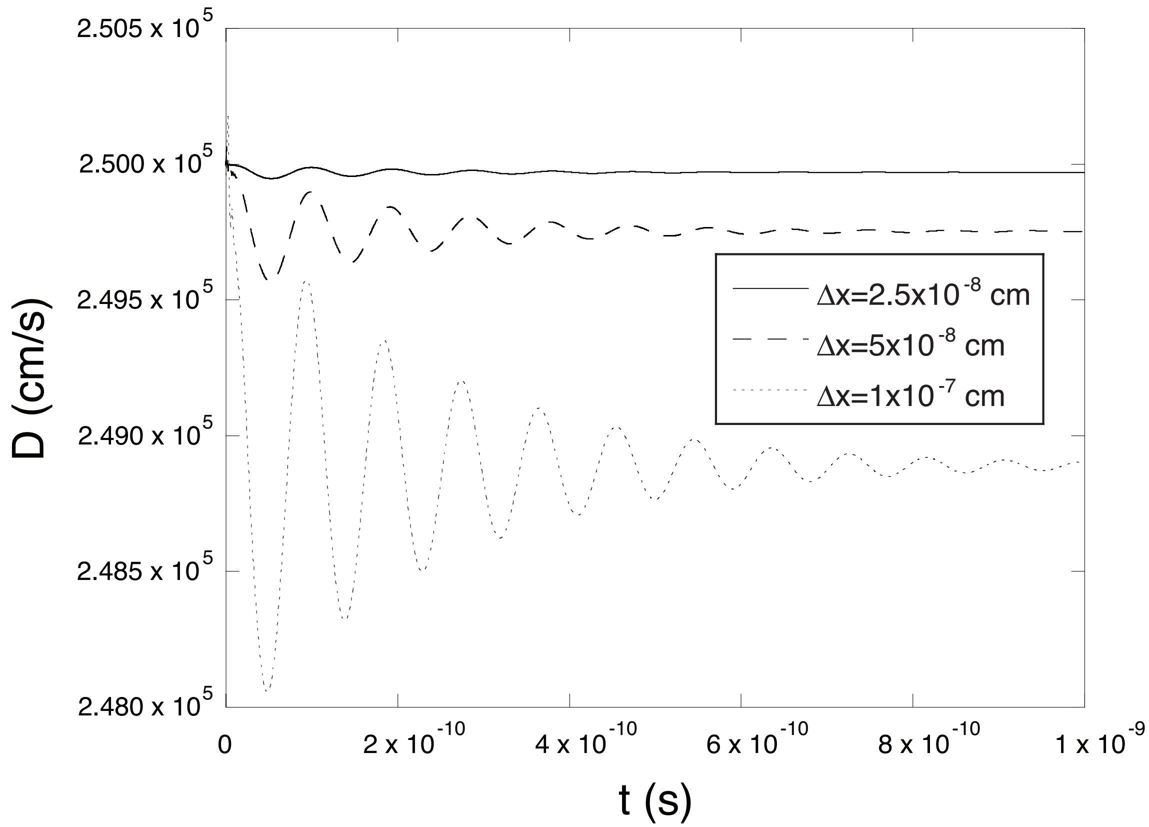
Stable Strongly Overdriven Case: Pressure

$D = 2.5 \times 10^5\ cm/s$.



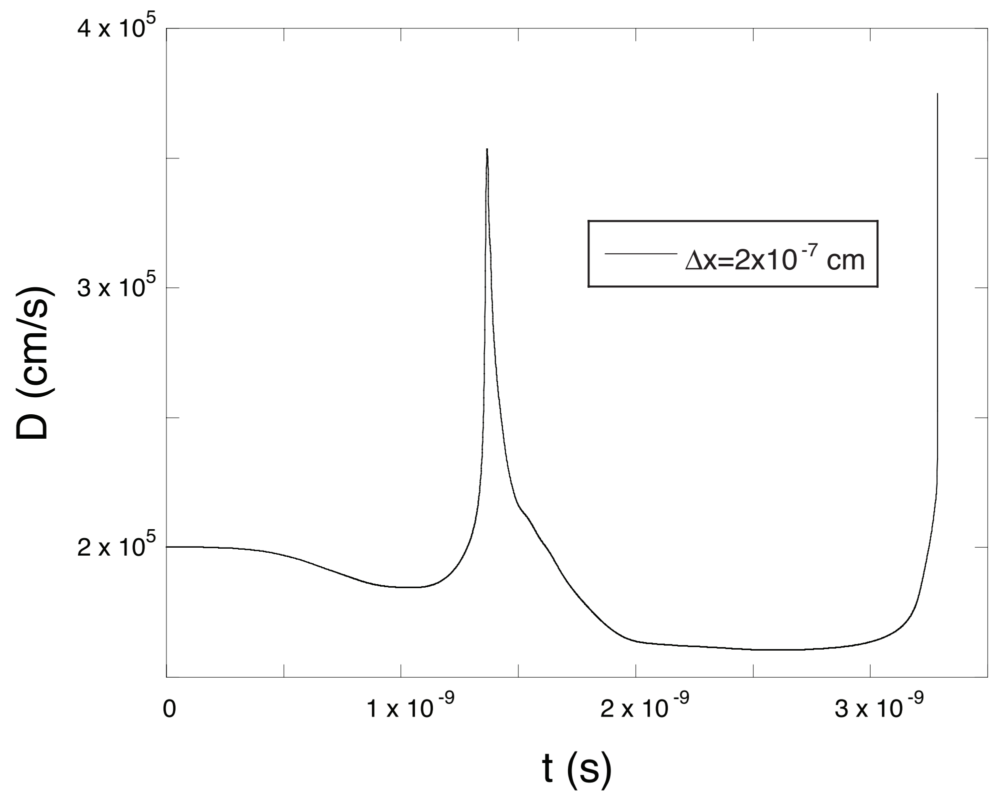
Stable Strongly Overdriven Case: Transient Behavior for various resolutions

Initialize with steady structure of $D = 2.5 \times 10^5\ cm/s$.



Unstable Moderately Overdriven Case: Transient Behavior

Initialize with steady structure of $D = 2 \times 10^5 \; cm/s$.

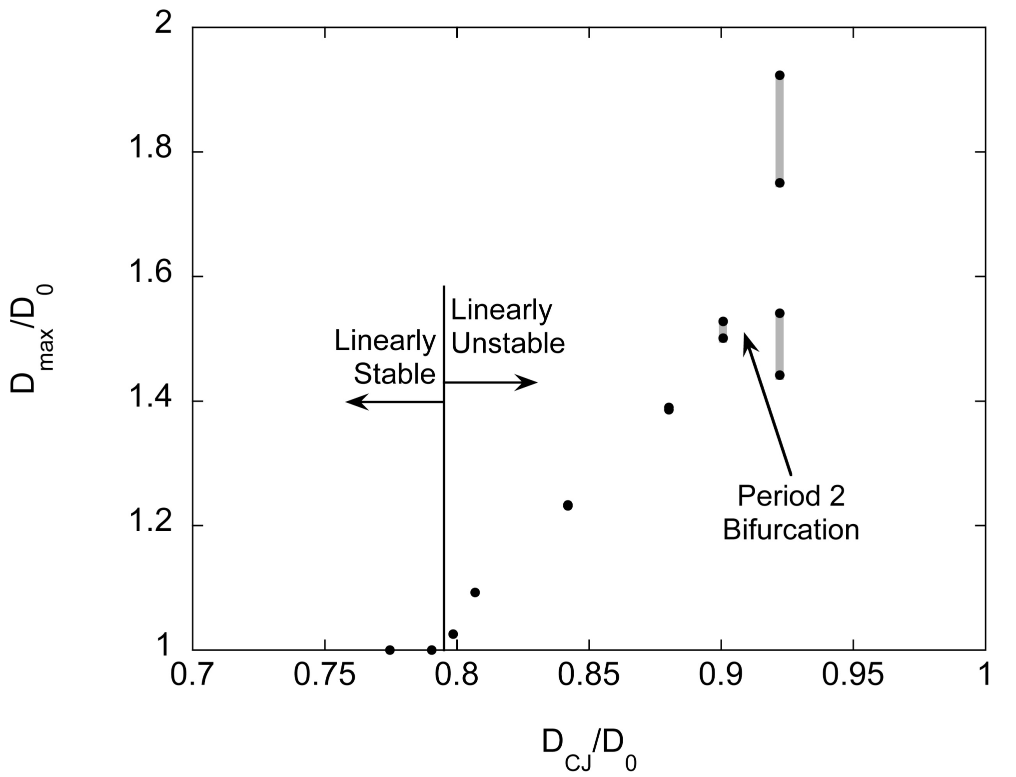


Effect of Resolution on Unstable Moderately Overdriven Case

Δx	Numerical Result
$1 \times 10^{-7}\ cm$	Unstable Pulsation
$2 \times 10^{-7}\ cm$	Unstable Pulsation
$4 \times 10^{-7}\ cm$	Unstable Pulsation
$8 \times 10^{-7}\ cm$	O_2 mass fraction > 1
$1.6 \times 10^{-6}\ cm$	O_2 mass fraction > 1

- Algorithm Failure for Insufficient Resolution
- At low resolution, one misses critical dynamics

Long Time Relative Maxima in D/D_o versus Inverse Overdrive



Diffusive Modeling in Gaseous Detonation

$$\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x}\left(\rho u\right) = 0,$$

$$\frac{\partial}{\partial t}\left(\rho u\right) + \frac{\partial}{\partial x}\left(\rho u^2 + p - \tau\right) = 0,$$

$$\frac{\partial}{\partial t}\left(\rho\left(e + \frac{u^2}{2}\right)\right) + \frac{\partial}{\partial x}\left(\rho u\left(e + \frac{u^2}{2}\right) + j^q + \left(p - \tau\right) u\right) = 0,$$

$$\frac{\partial}{\partial t}\left(\rho Y_B\right) + \frac{\partial}{\partial x}\left(\rho u Y_B + j_B^m\right) = \rho r,$$

Diffusive Modeling in Gaseous Detonation

$$
\begin{aligned}
p &= \rho R T, \\
e &= c_v T - q Y_B = \frac{p}{\rho\,(\gamma - 1)} - q Y_B, \\
r &= H(p - p_s) a\,(1 - Y_B)\, e^{-\frac{E}{p/\rho}}, \\
j_B^m &= -\rho \mathcal{D} \frac{\partial Y_B}{\partial x}, \\
\tau &= \frac{4}{3} \mu \frac{\partial u}{\partial x}, \\
j^q &= -k \frac{\partial T}{\partial x} + \rho \mathcal{D} q \frac{\partial Y_B}{\partial x}.
\end{aligned}
$$

To compare with previous one-step work...

- Need to choose scale ratios between diffusion and reaction
- Choose half-reaction length scale to be $1 \mu m$.
- Choose diffusive length scale to be $100 nm$

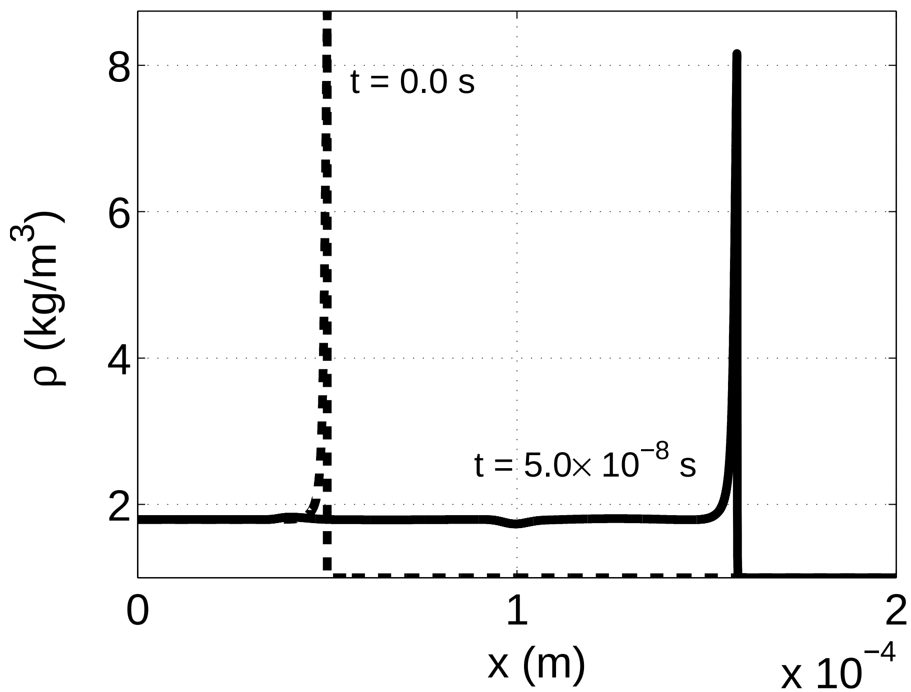
$$\mathcal{D} = 10^{-4}\, m^2/s,$$
$$k = 10^{-1}\, W/m/K,$$
$$\mu = 10^{-4}\, Ns/m^2,$$

For $\rho_o = 1\, kg/m^3$, $Le = Sc = Pr = 1$.

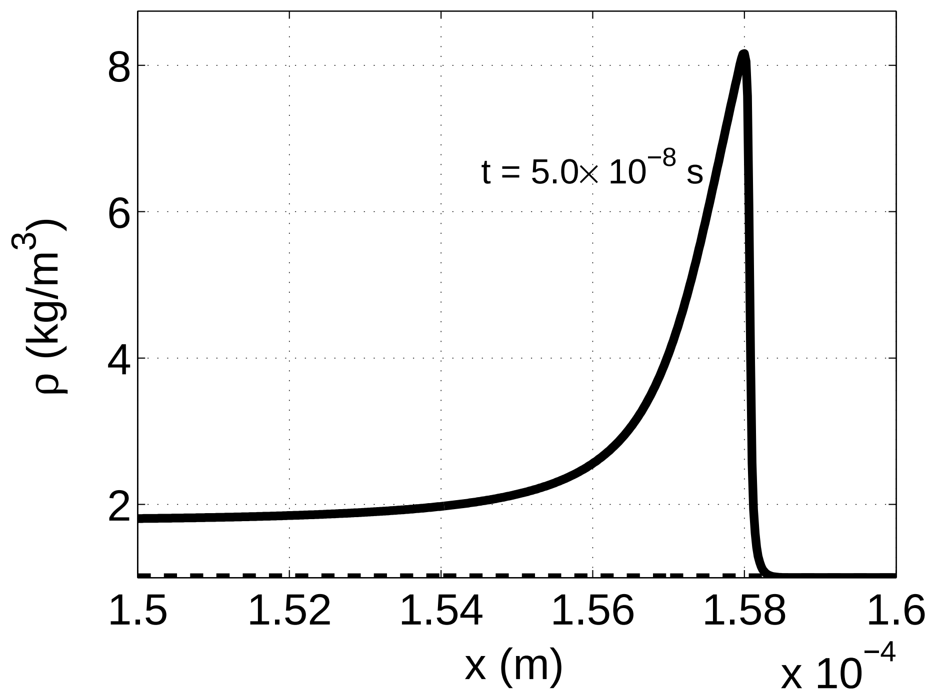
Numerical Methods

- 5th Order WENO Schemes for Hyperbolic Components
- 4th Order Central Difference Scheme for Parabolic Components
- 3rd Order Explicit Runge-Kutta Time Integration
- Expect Fully 4th Order Convergence Rates Under Resolution

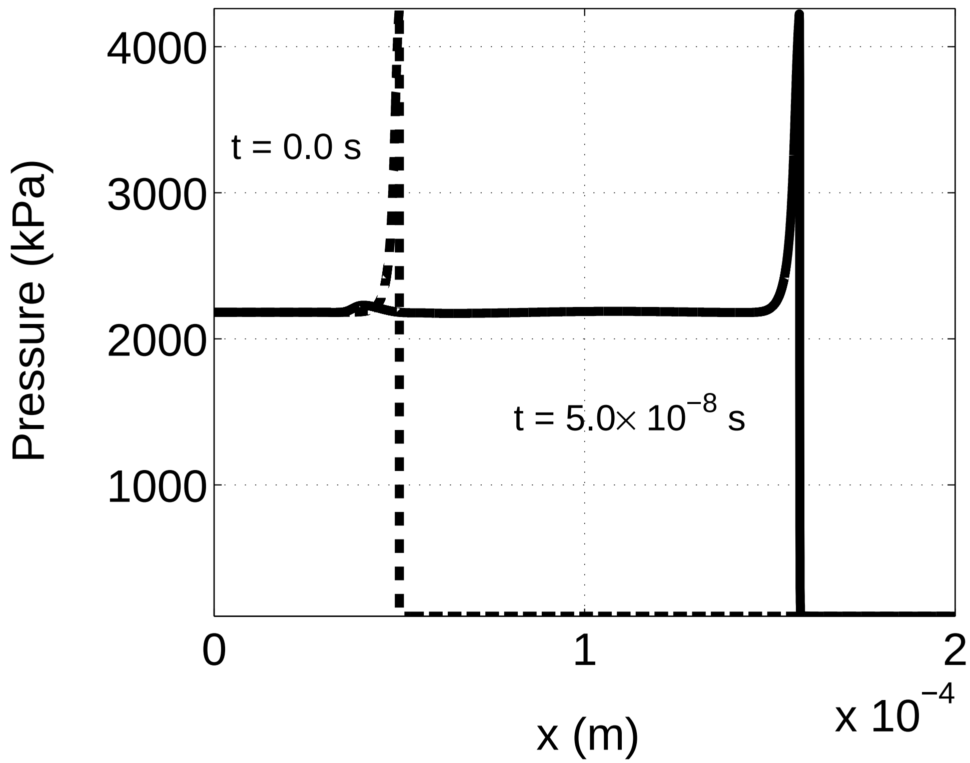
Density for a stable detonation $E = 25$



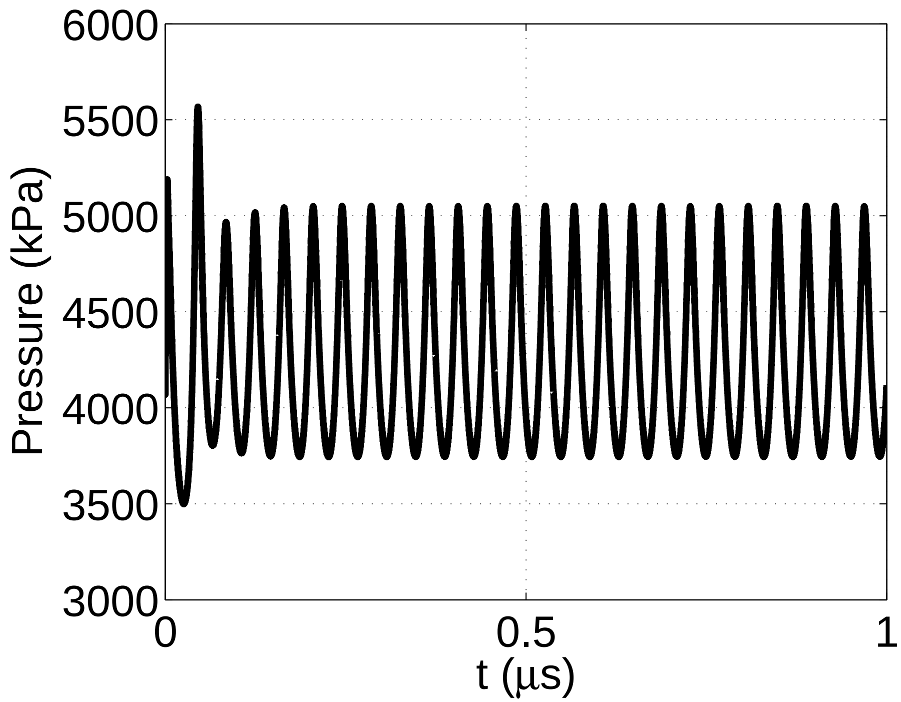
Density for a stable detonation $E = 25$ - zoom



Pressure for a stable detonation $E = 25$ - zoom

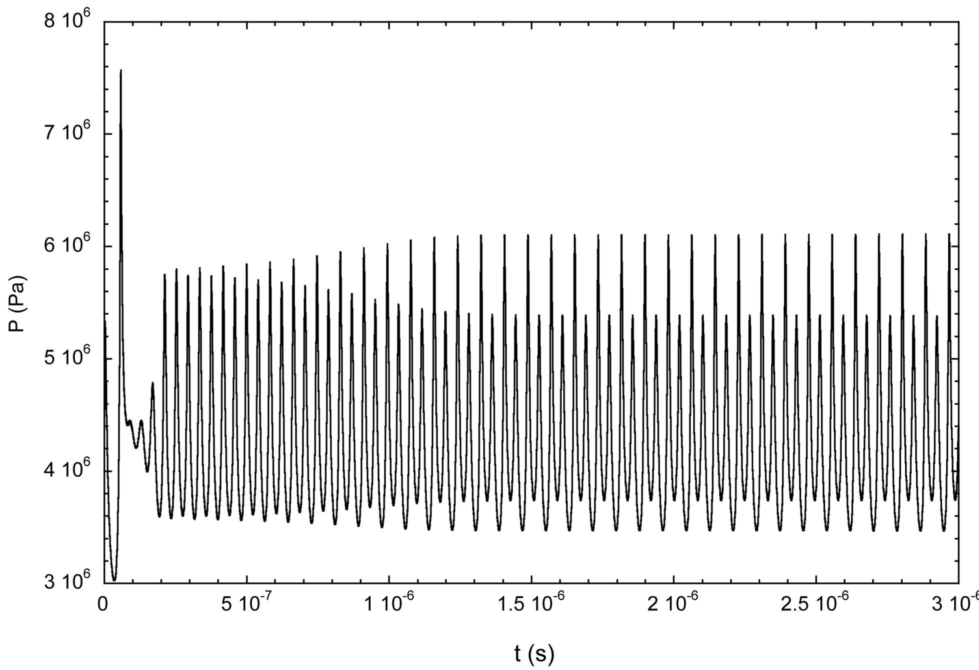


Pressure vs. time for a unstable detonation $E = 28$



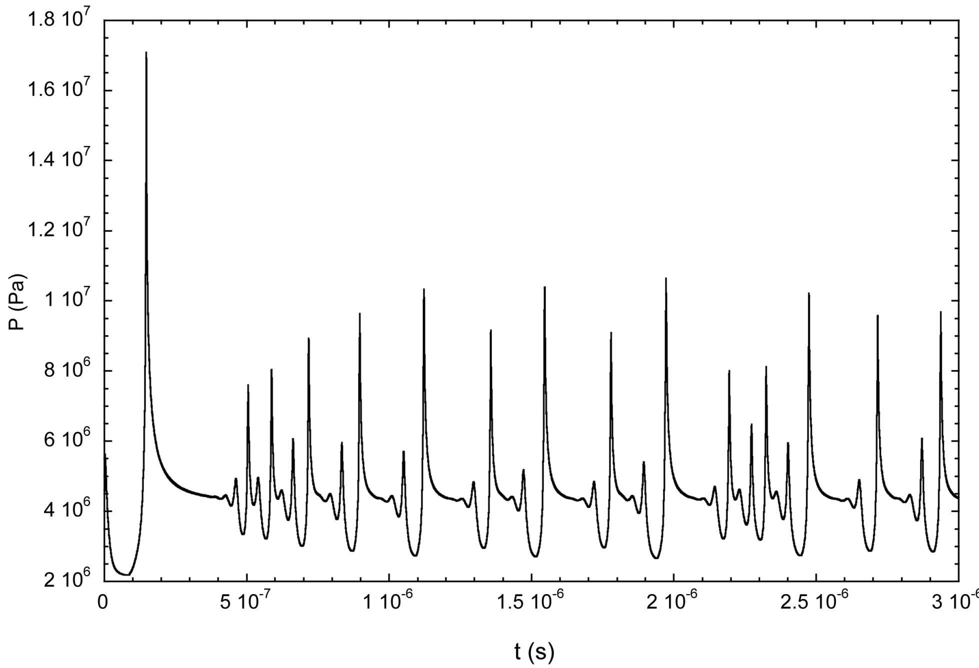
Pressure vs. time for a unstable detonation

$$E = 29.5$$



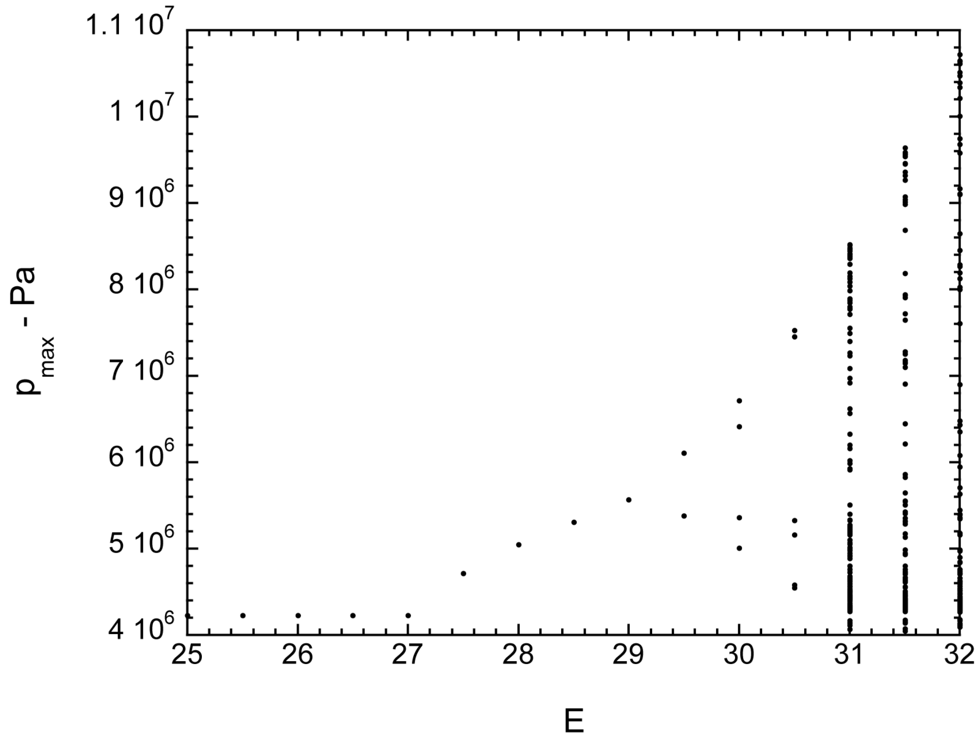
Period Doubling

Pressure vs. time for a unstable detonation $E = 32$



Chaotic Dynamics

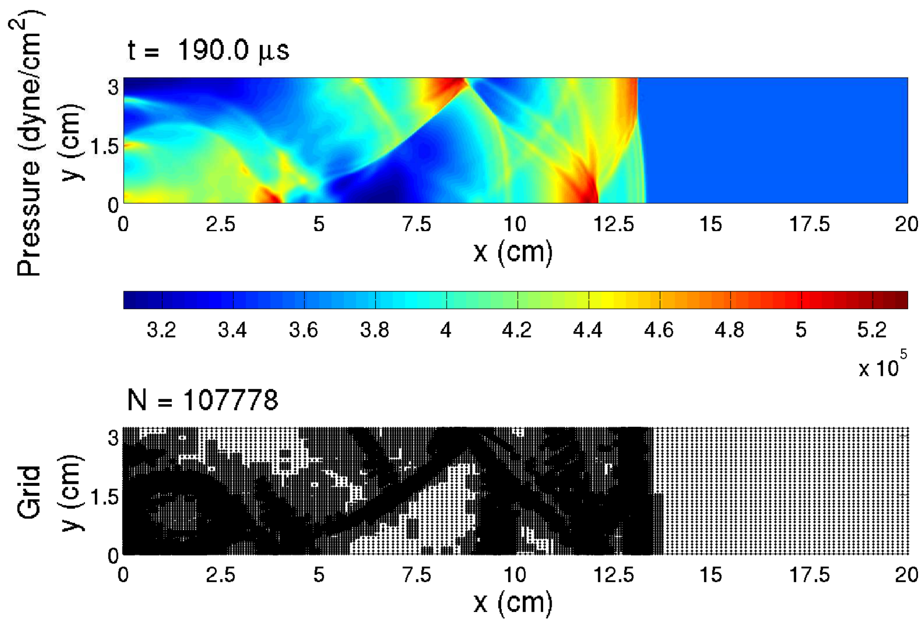
Diffusive Bifurcation Diagram



With a relatively small amount of diffusion, a substantial stabilization occurs.

Where we are headed with all this...

Multi-D WAMR simulation of $2H_2 : O_2 : 7Ar$



Conclusions

- Unsteady detonation dynamics can be accurately simulated when sub-micron scale structures admitted by detailed kinetics are captured with ultra-fine grids.
- Shock fitting coupled with high order spatial discretization assures numerical corruption is minimal.
- For resolved diffusive effects, relatively simple numerical methods work fine.
- Predicted detonation dynamics consistent with results from inviscid models...
- At these sub-micron length scales, diffusion plays a substantial role.