Instabilities in the Flow of Thin Liquid Films

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Overview

- Examples of some current applications
- General physical and mathematical issues related to thin film flows
- Review of flows involving contact lines
- Examples of (mostly) computational results involving dynamics of thin films
  - Instability development and pattern formation
  - Flows on inhomogeneous surfaces
  - Issues related to partial wetting

In collaboration with Javier Diez and other members of the Fluids group at FCEx UNCPBA Tandil: Alejandro Gonzalez, Roberto Gratton, Juan Gomba

Examples of previous works

- Inclined plane flow
  - Troian, Europhys. Lett. '89
  - Lopez etal JFM '96
  - Eres etal PoF '00
  - Perazzo & Gratton PoF '04
- Spinning drops
  - Spaid & Homsy, J. Non-Newt. Fluid Mech '94
- Thermally driven flows
  - Cazabat etal, Nature '90
  - Sur etal PRL '03
- Flows with surfactants
  - Matar & Kumar SIAM J. Appl. Math '04
- Two layer flows
  - Thiele et al PRE '04
  - Segin etal JFM '05

Numerous applications

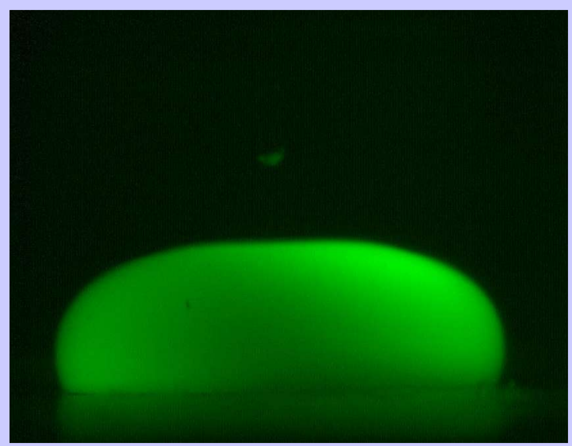
- Spin coating
- Photographic films
- Microscopic fluid devices (MEMS, etc.)
- Spraying

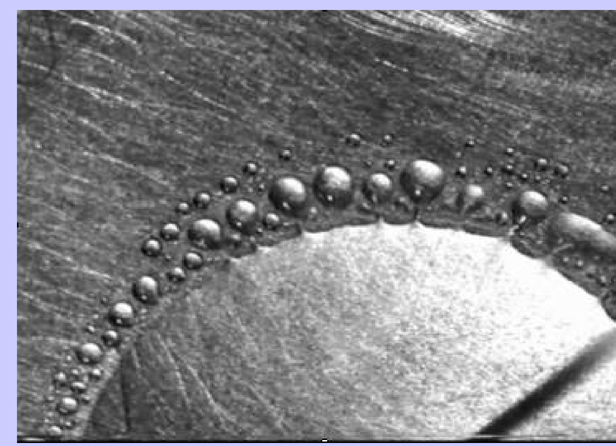
- Some examples of recent experiments showing dynamics of thin films and drops

Experiments involving electrowetting

- Apply electric field and modify fluid wetting properties (contact angle change)

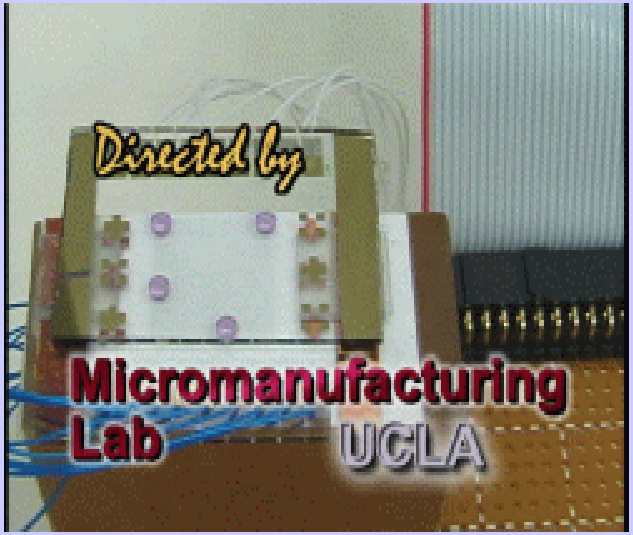
F. Mugele's group at Twente University





More about electrowetting

C. J. Kim's group at UCLA



R. Fair's group at Duke

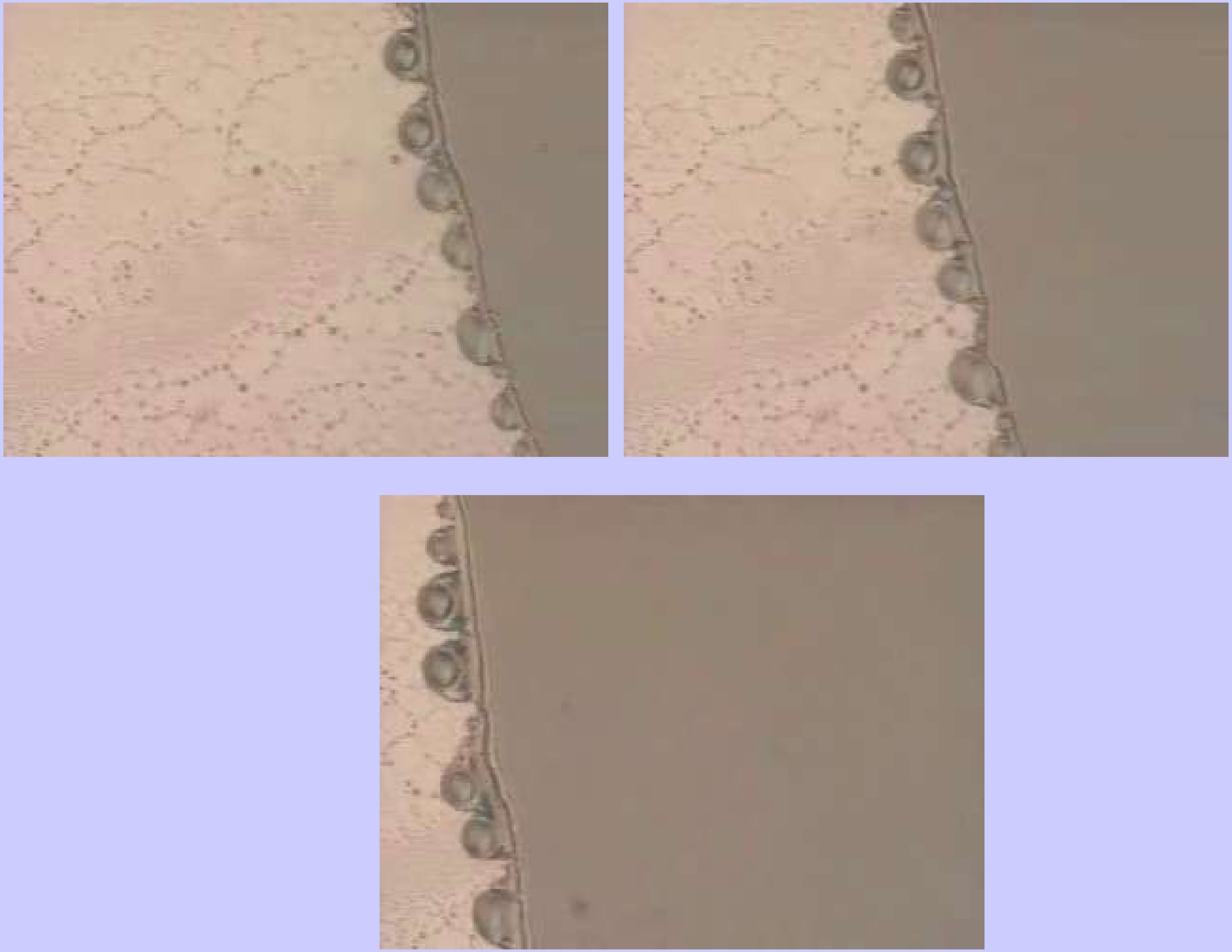
Flows that include thermal effects

- Example from industrial world: flow on extremely clean silicon wavers

  By Y. Gotkis, KLA-Tencor, San Jose CA

  - Peculiar droplet(s) ejection from evaporative mother-drop
  - Relevance: numerous processes in semiconductor industry involving processes on micro- and nano scale
  - Flows of alcohols and alcohol-water mixtures

Alcohol-water mixture: formation of convection rolls at the fronts



Octopi-like features detach from the fluid front (pure alcohol)

Challenges

- Understand the details of the physics at the contact line
- Extend to complex fluids
- Properly account for additional forces (electrowetting, thermal effects, evaporation, ...)
- Bridge the scales: from micro to meso to macro
- Compute accurately the flow: multiscale problem

Plenty of room for new physics!!!

From Navier-Stokes to a single PDE

- Free surface flows are very difficult to address due to continuously changing domain of interest
- Need to simplify the formulation as much as possible
  - Use the fact that the films are thin, fluids are incompressible and Newtonian
  - Ignore thermal effects and evaporation
  - Assume simple models for fluid-solid interaction

Assumptions

- Fluid is thin and all gradients are small
- Inertial effects can be ignored
- Capillary number is small
- No-slip boundary condition at liquid-solid interface (to be discussed)
- Consider first completely wetting fluids; extend later to partial wetting

Reduction (1)

Navier – Stokes equations:

$$\frac{\partial \boldsymbol{u}}{\partial t} + (\boldsymbol{u} \cdot \nabla)\boldsymbol{u} = -\frac{1}{\rho}\nabla p + \frac{\mu}{\rho}\nabla^2 \boldsymbol{u} + g\sin(\alpha)\boldsymbol{i} - g\cos(\alpha)\boldsymbol{k}$$

$$\boldsymbol{u} = (\boldsymbol{v}, w)$$

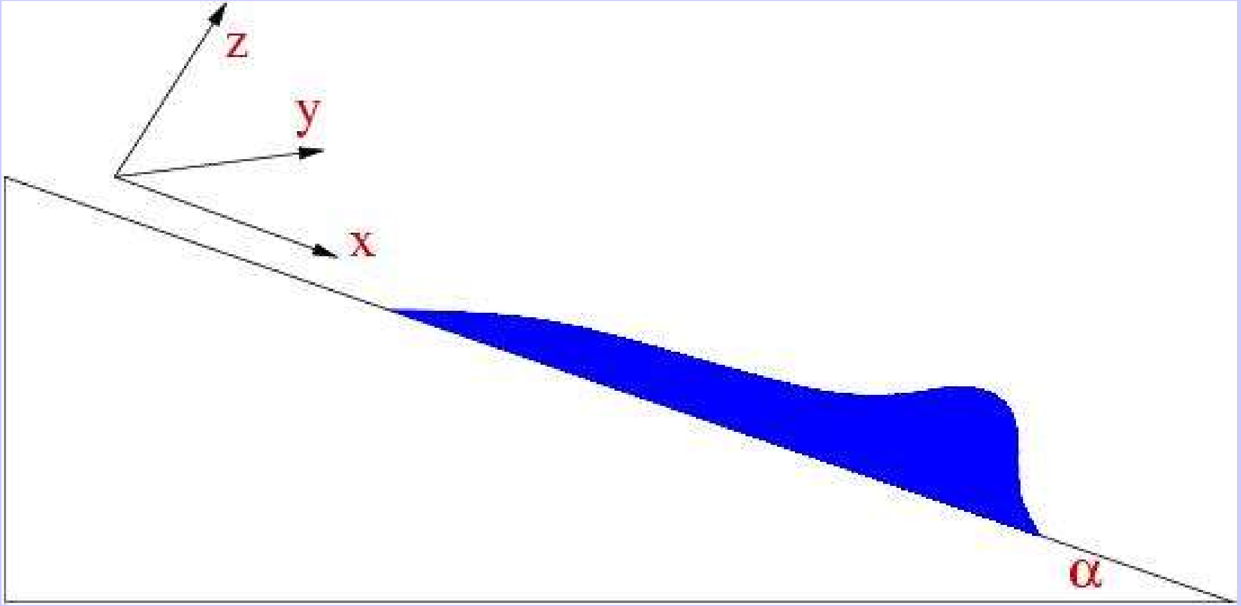
$$\longrightarrow \frac{\partial^2 \boldsymbol{v}}{\partial z^2}$$

use incompressibility

$$\nabla \cdot \boldsymbol{u} = 0$$

to show that

$$w \ll |\boldsymbol{u}|$$



Reduction (2)

$x - y$ components

$$\nabla_2 p = \mu \frac{\partial^2 \boldsymbol{v}}{\partial z^2} + \rho g \sin(\alpha) \boldsymbol{i} \qquad (1)$$

z component

$$\frac{\partial p}{\partial z} = -\rho g \cos(\alpha) \qquad (2)$$

Use Laplace – Young condition $p \sim -\gamma \kappa \quad at \quad z = h(x, y)$
to solve (2)

$$p = -\rho g (z - h) \cos(\alpha) - \gamma \kappa + const.$$

Integrate (1) twice using

no slip at fluid solid interface $\quad |\boldsymbol{v}| = 0 \quad at \quad z = 0$

continuity of stresses at fluid-air interface $\quad \left| \frac{\partial \boldsymbol{v}}{\partial z} \right| = 0 \quad at \quad z = h(x, y)$

Reduction (3)

equation for the fluid velocity

$$\boldsymbol{v}=\frac{1}{\mu}\left[\nabla_2\left(\rho g h \cos(\alpha)-\gamma\kappa\right)-\rho g \sin(\alpha)\boldsymbol{i}\right]\left[\frac{z^2}{2}-hz\right]$$

Average over fluid thickness

$$\langle\boldsymbol{v}\rangle=\frac{1}{h}\int_0^h \boldsymbol{v}\,dz$$

Approximate curvature of the fluid-air interface

$$\kappa\approx\nabla^2 h$$

Use mass-conservation

$$\frac{\partial \boldsymbol{v}}{\partial t}+\nabla\cdot\left(h\langle\boldsymbol{v}\rangle\right)=0$$

to obtain

Thin film equation

Fourth order nonlinear PDE for the fluid height:

$$3\mu\frac{\partial h}{\partial t}+\nabla\cdot[\gamma h^3\nabla\nabla^2 h]-\rho g\cos(\alpha)\nabla\cdot[h^3\nabla h]+\rho g\sin(\alpha)\frac{\partial h}{\partial x}=0$$

capillarity

out-of-plane gravity

in-plane gravity

h: fluid thickness

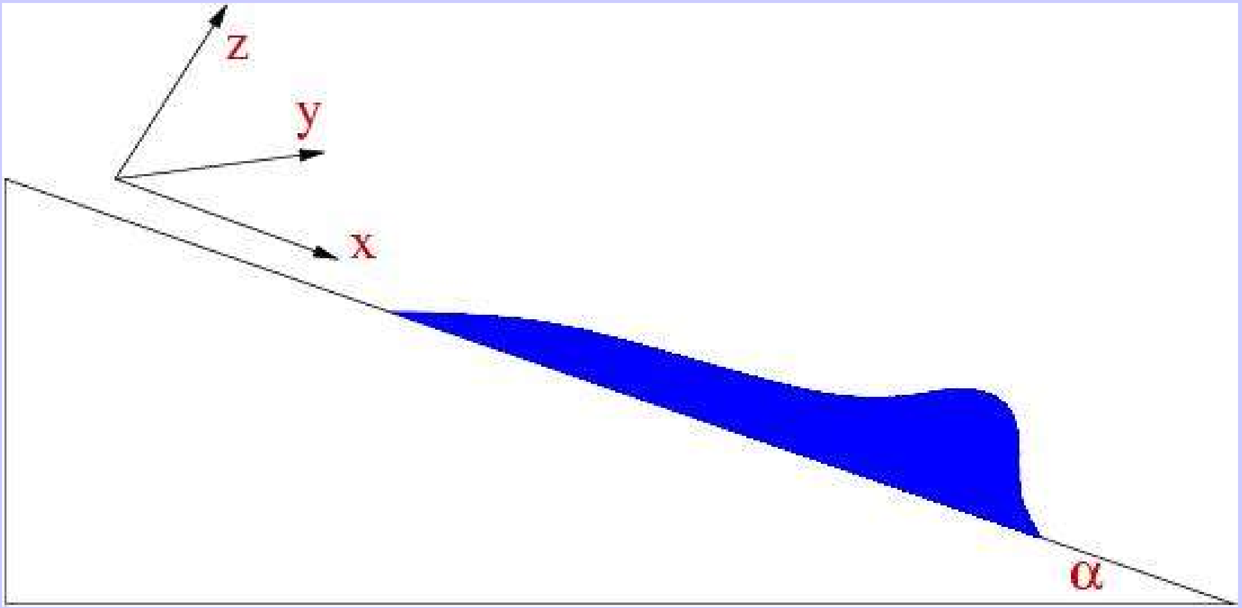
α: inclination angle

ρ : fluid density

μ : fluid viscosity

γ : surface tension

g : gravity



Scales

- scale fluid thickness by H: thickness far from the contact line
- in-plane length scale : l
- velocity scale: U
- time scale: $t = l/U$
- Capillary number: $Ca=\mu \frac{U}{\gamma}$

Non-dimensional equation:

$$\frac{\partial h}{\partial t}+\nabla \cdot [h^3 \nabla \nabla^2 h]-D(\alpha)\nabla \cdot [h^3 \nabla h]+\frac{\partial h}{\partial x}=0$$

$$D(\alpha)=(3\,Ca)^{1/3}\cot(\alpha)$$

Contact line singularity

- Add precursor film of thickness b
  - spreading on a prewetted surface
  - introduces new length-scale

    Dussan & Davis, JFM '74

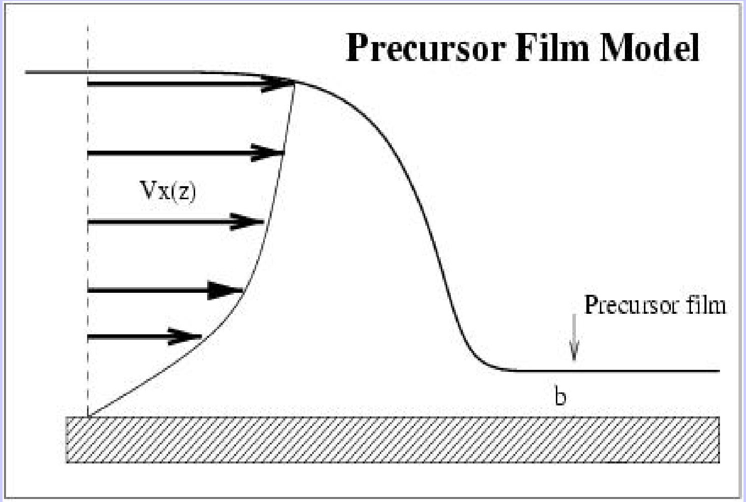
    de Gennes, RMP '85

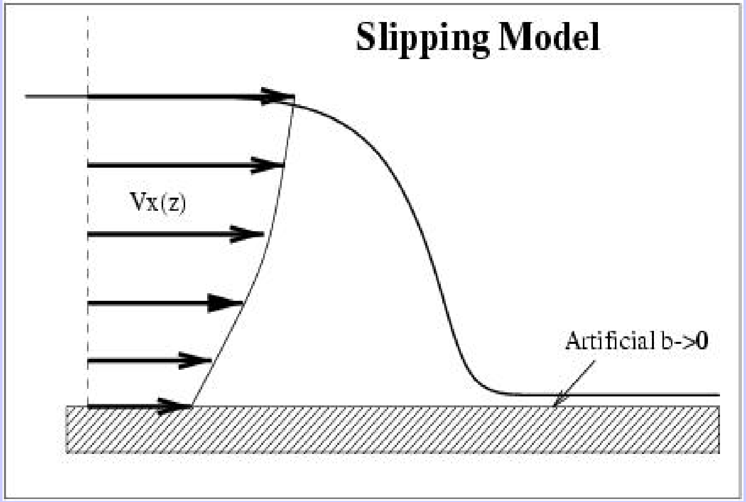
    Goodwin & Homsy, PoF A '91

    Gonzalez etal PRE '04
- Relax no-slip boundary condition

- For the purpose of understanding macro-behavior, the models are consistent
- For the purpose of computing, precursor model is more efficient

  Diez, Kondic, Bertozzi, PRE '01





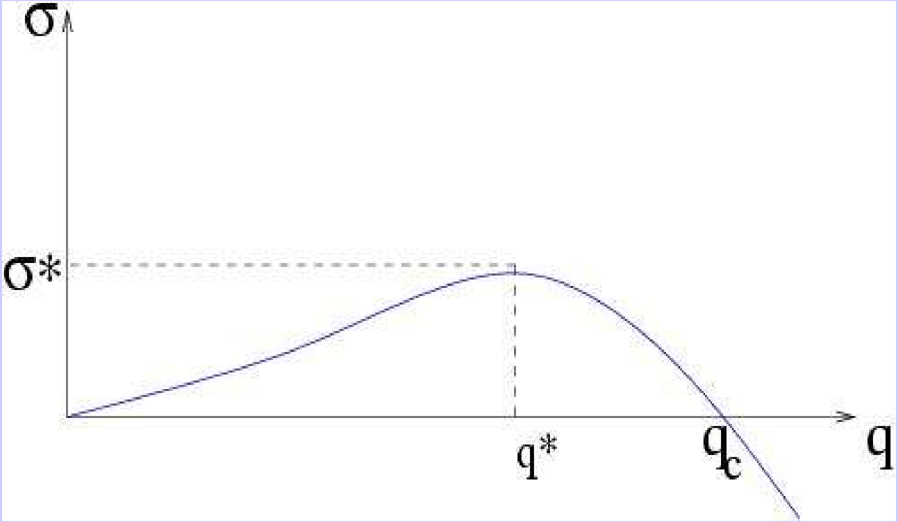
Linear stability analysis (flow down an incline)

- Expand about *y*-independent solution (trivial traveling wave)

$$h(x,y,t)=h_0(x-Vt)+e^{\sigma t}e^{iqy}h_1(x)$$

There is a critical wave number q_c below which the flow is unstable

$$\left(\lambda_c=2\pi/q_c\approx 8\right)$$



Troian, Europhys. Lett. '89
Bertozzi & Brenner PoF '97

Computational methods

- Very demanding problem due to high degree, nonlinearity and presence of short scales (precursor)
- Efficiency, accuracy and stability are crucial even with state-of-the-art computers
  - Finite difference methods
    - ADI methods
      - Eres etal PoF '00, Witelski Appl. Num. Math. '03
    - Fully implicit methods
      - Diez & Kondic PRL '01, PoF '02
  - Spectral and pseudospectral methods
      - Thiele etal PRE '01 etc

Implemented computational methods

- Implicit time discretization
- Time step: accuracy requirement
- Spatial linearization: Newton method
- Iterative biconjugate gradient solver for linear problem
- Second order in space and time
  - Details: Diez & Kondic, JCP '02

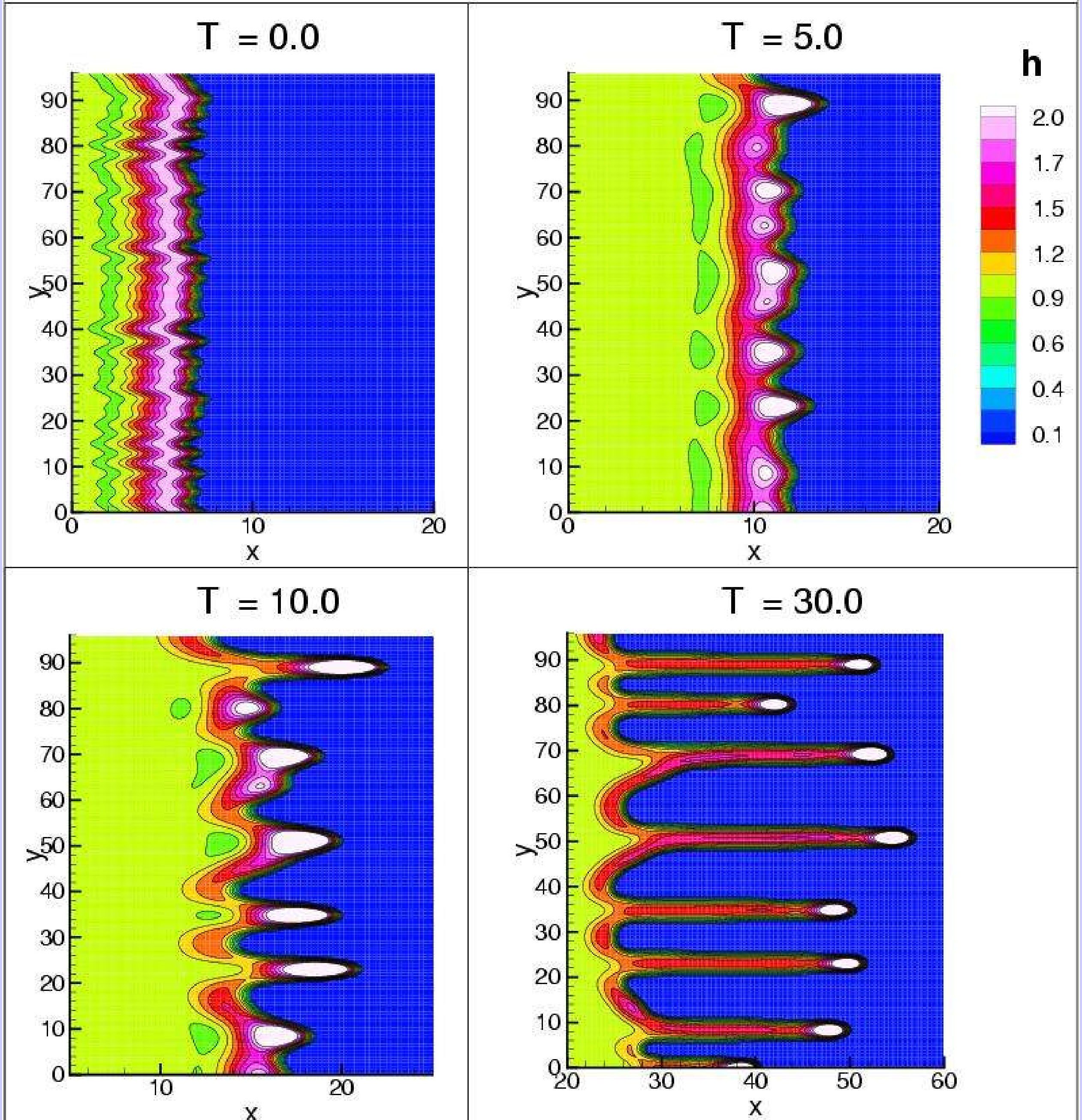
Initial condition: 1D profile modified by harmonic perturbations:

$$A(t=0)=\sum_i^N A_i \cos(q_i y); \qquad A_i \in [-0.1, 0.1]$$

FLOW DOWN A VERTICAL PLANE

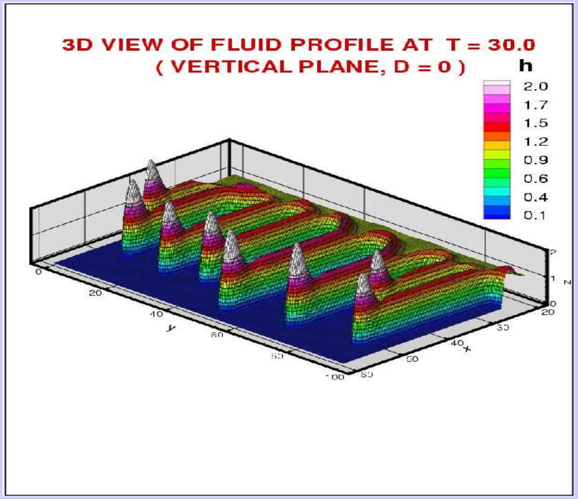
$b = 10^{-2}$ $dx = 0.2$ $L_x = 20 - 40$ $\lambda_i = 2\,L_y/\,i,\ \ i=1,....,50$

$dy = 0.5$ $L_y = 96$ $A_{max} = 0.1$ (random)



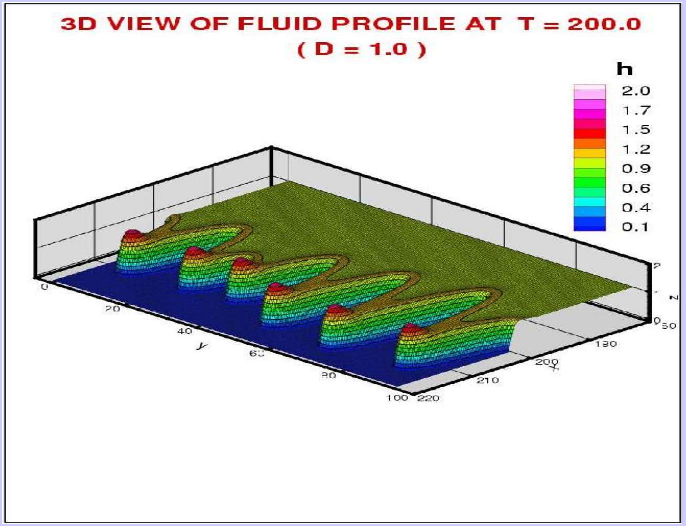
Pattern formation – vertical plane

- Stable (short) wavelengths disappear for very early times (coarsening)
- Emerging wavelengths center about $\lambda^* = 2\pi/q^*$ predicted by linear theory
- Occurrence of a range of wavelengths, as in experiments
- Formation of finger-like patterns



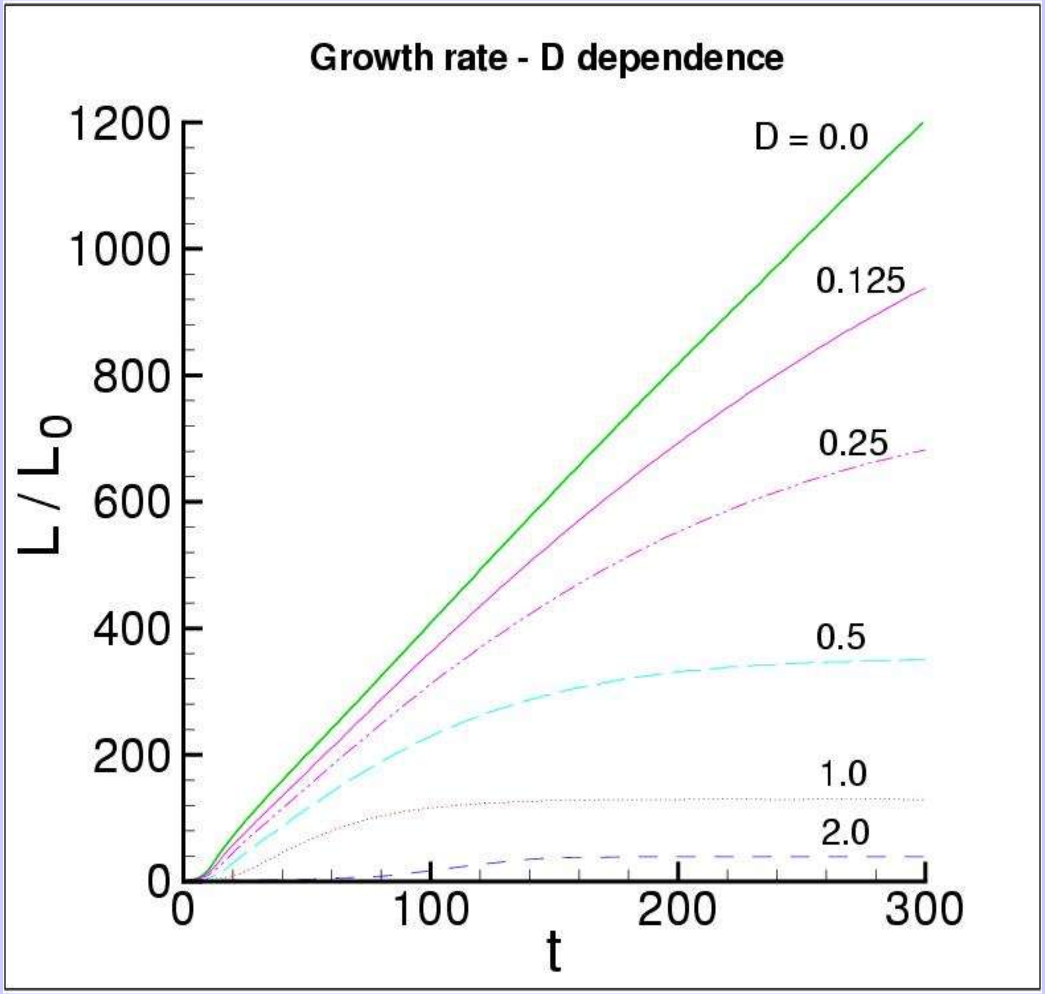
Pattern formation – inclined plane

- Longer wavelengths for smaller angles
- Significant modification of emerging patterns: transition from fingers to saw-tooth patterns
  - Diez & Kondic, PRL '01
- Good quantitative agreement with experiments
  - Johnson etal JFM '99
- Question: do the patterns grow for all times?



Pattern growth

- Perturb the fluid by initial perturbation L_0 and record the pattern length $L(t)$
- *D = 0:* linear growth even for long times
- *D > 0:* growth stops



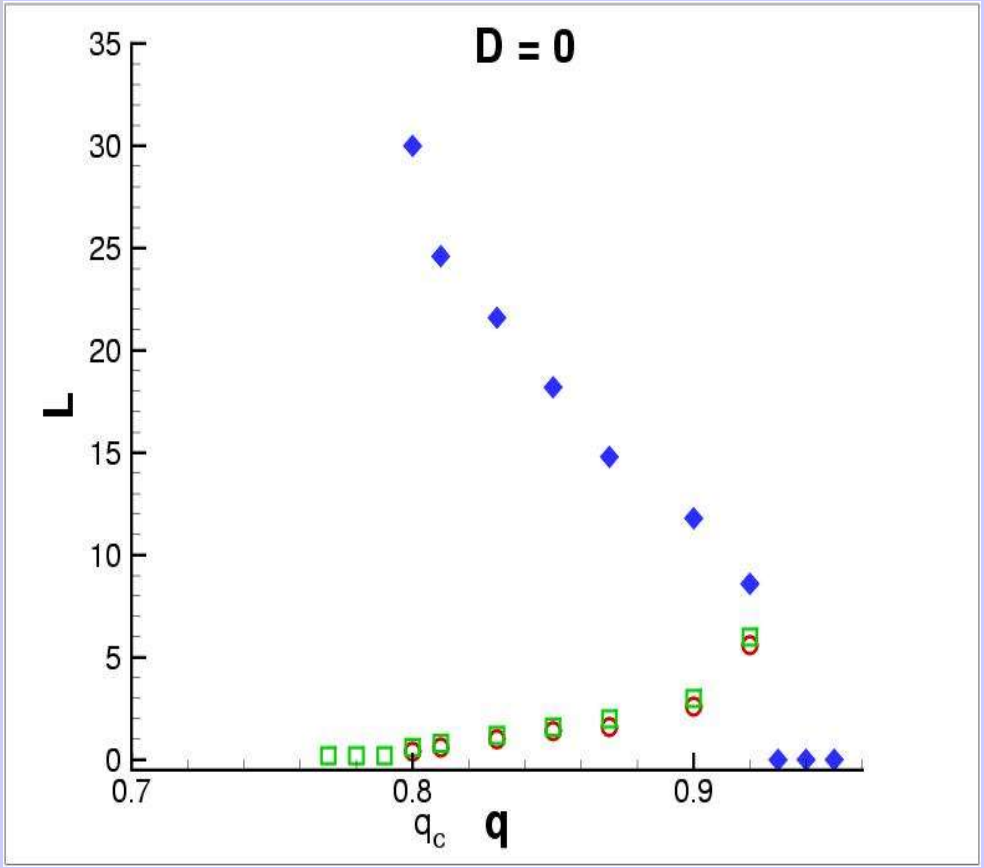
Existence of nontrivial traveling wave solutions!!!

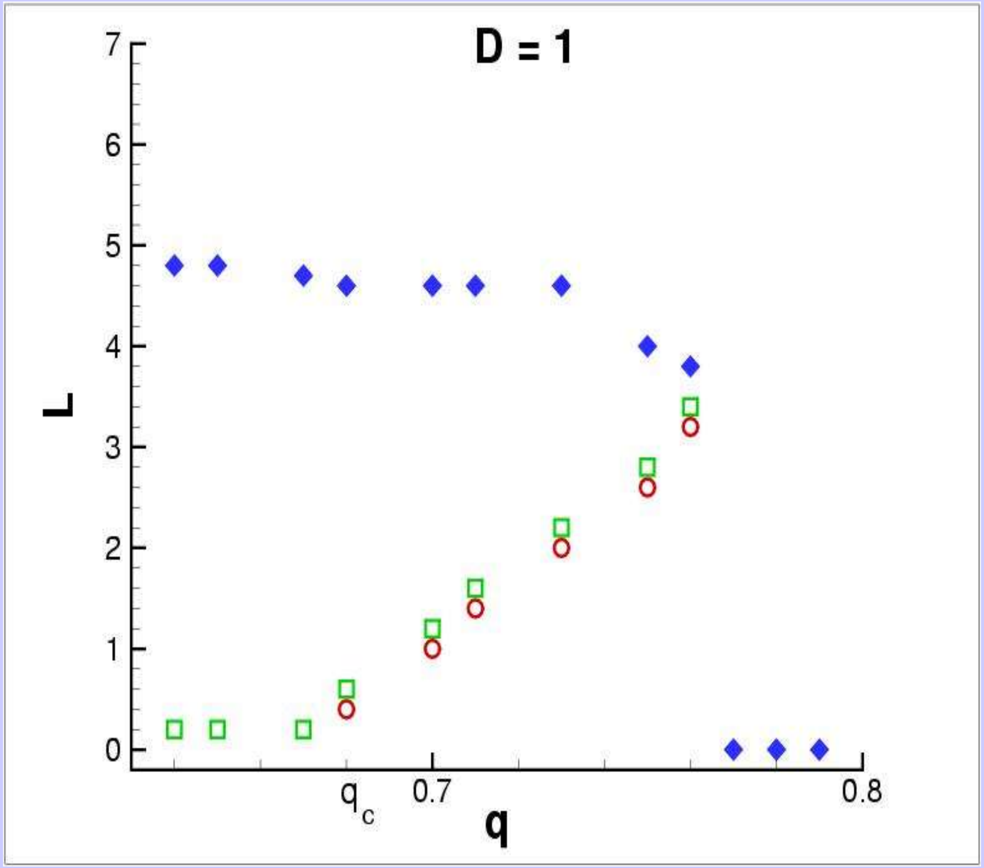
Growth saturation

- Relevance: numerous applications in which pattern growth is crucial for performance (coatings, etc)
- Challenges: perform accurate long time simulations using reasonably realistic value of precursor film thickness; avoid spurious saturation effects due to large precursor and/or small computation domain
  - See Eres et al. PoF '00

Bifurcation analysis

- Analyze the influence of domain size and initial perturbation amplitude for $D \geq 0$

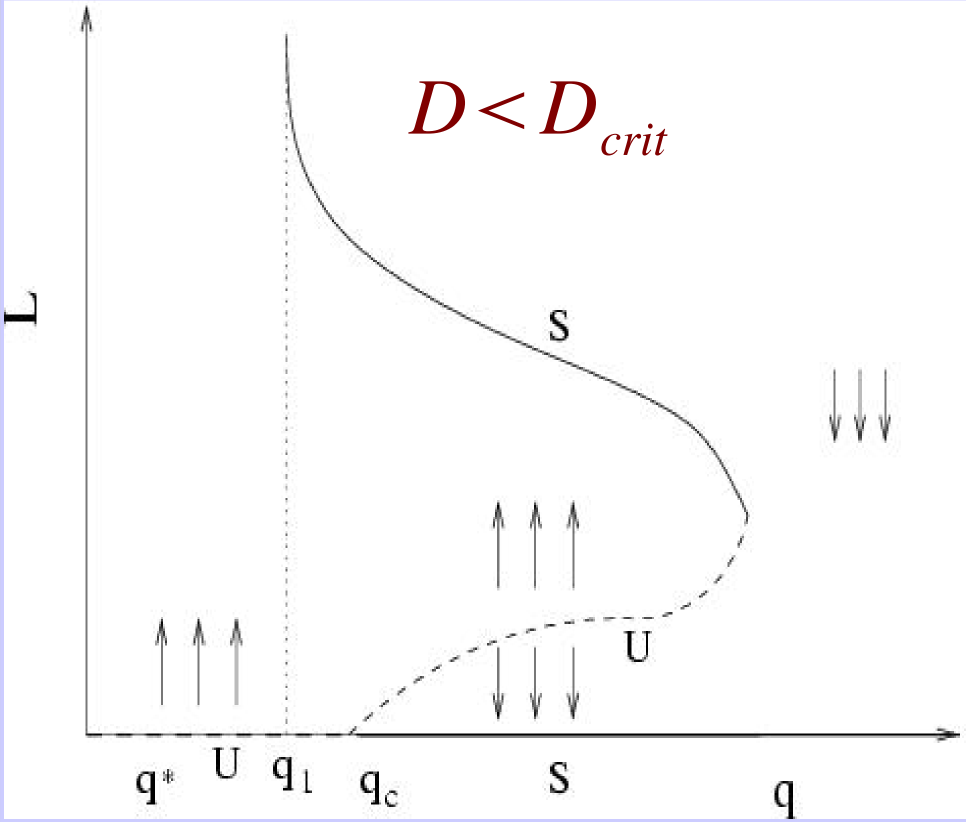


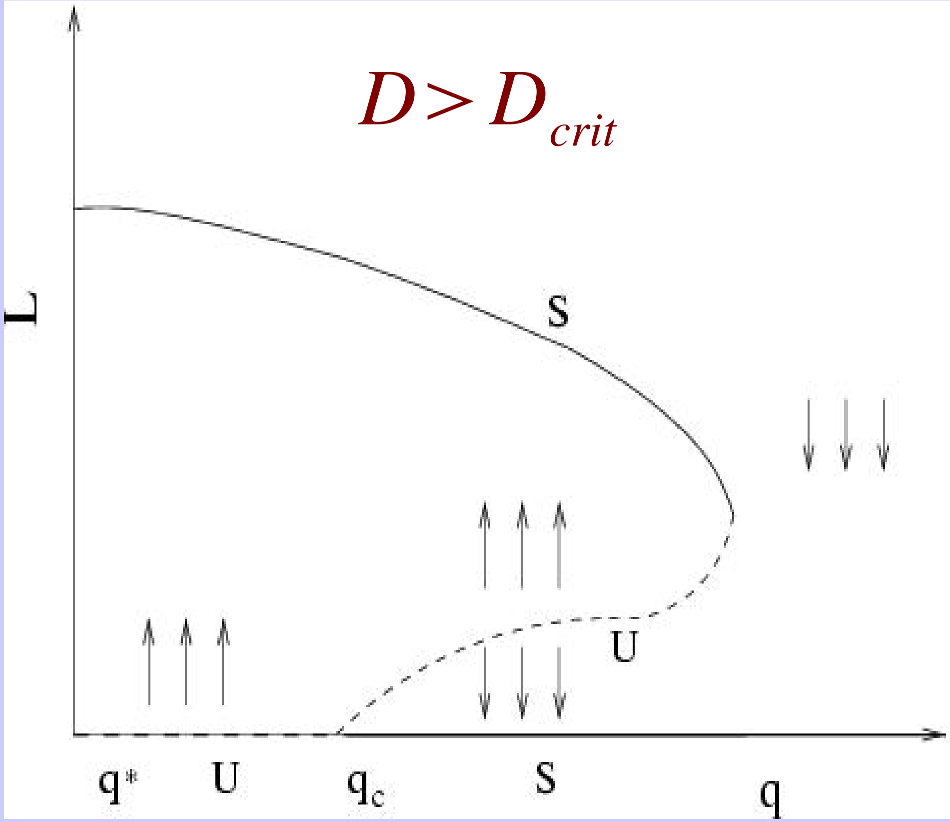


- See Kondic & Diez, Physica D '05

Bifurcation analysis: results and questions

- Very different behavior for $D<D_{crit} \wedge D>D_{crit}$
- Is $D_{crit}=0\,?$





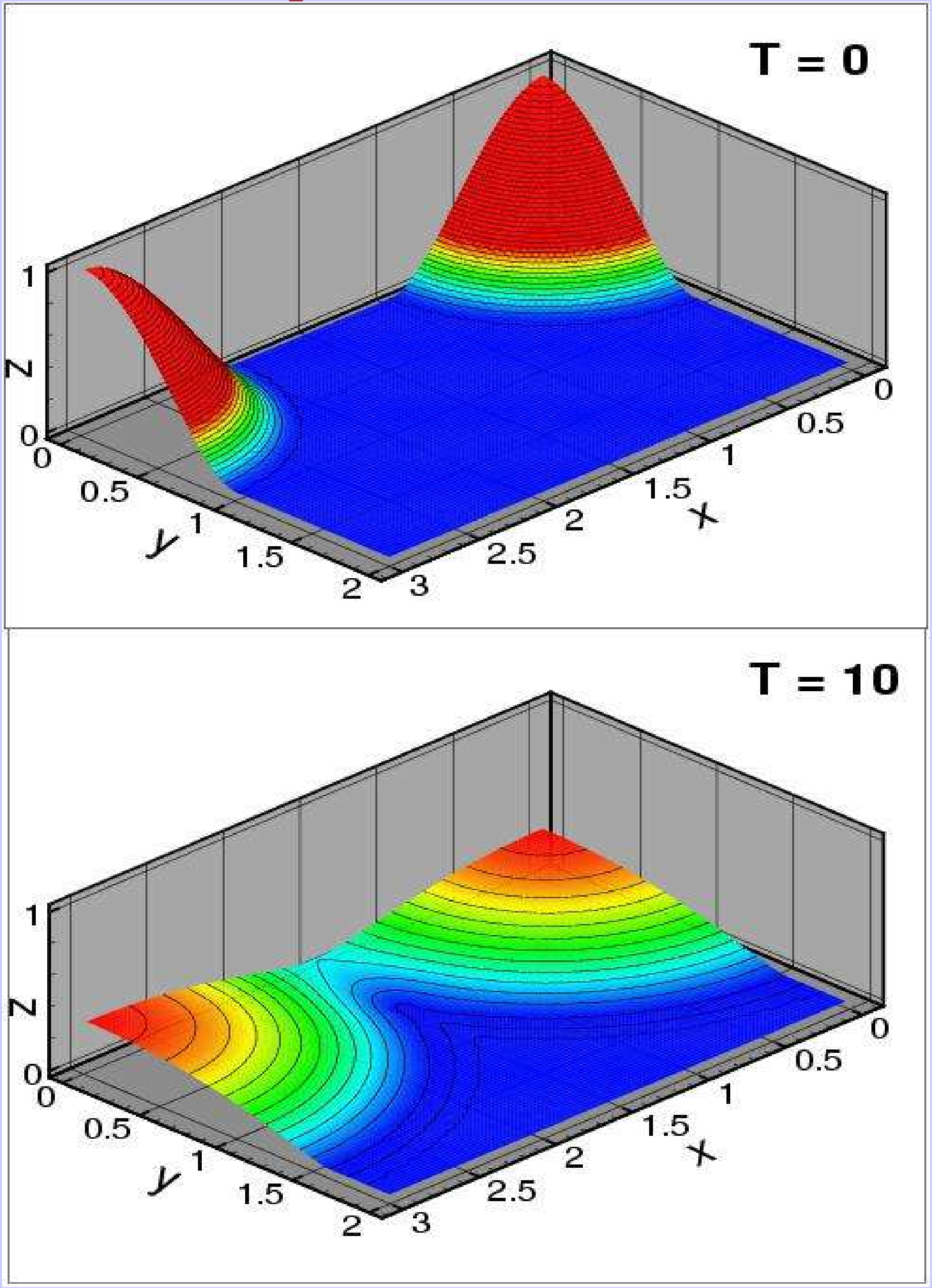
Can traveling wave solutions be found analytically?

Application: Coalescence of Sessile Drops

- Example of a problem where calculations on nonuniform grids were crucial
  - Diez & Kondic, JCP '02
- Applications: coating, spraying
- Follow spreading through `topological' tran-sition as drops merge

Merge of two drops

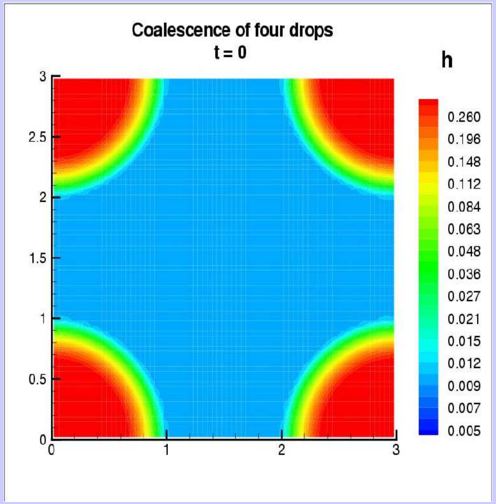
- Use symmetry of the problem
- Analyze some peculiar flow features
- Discuss front motion in comparison to single drop spreading

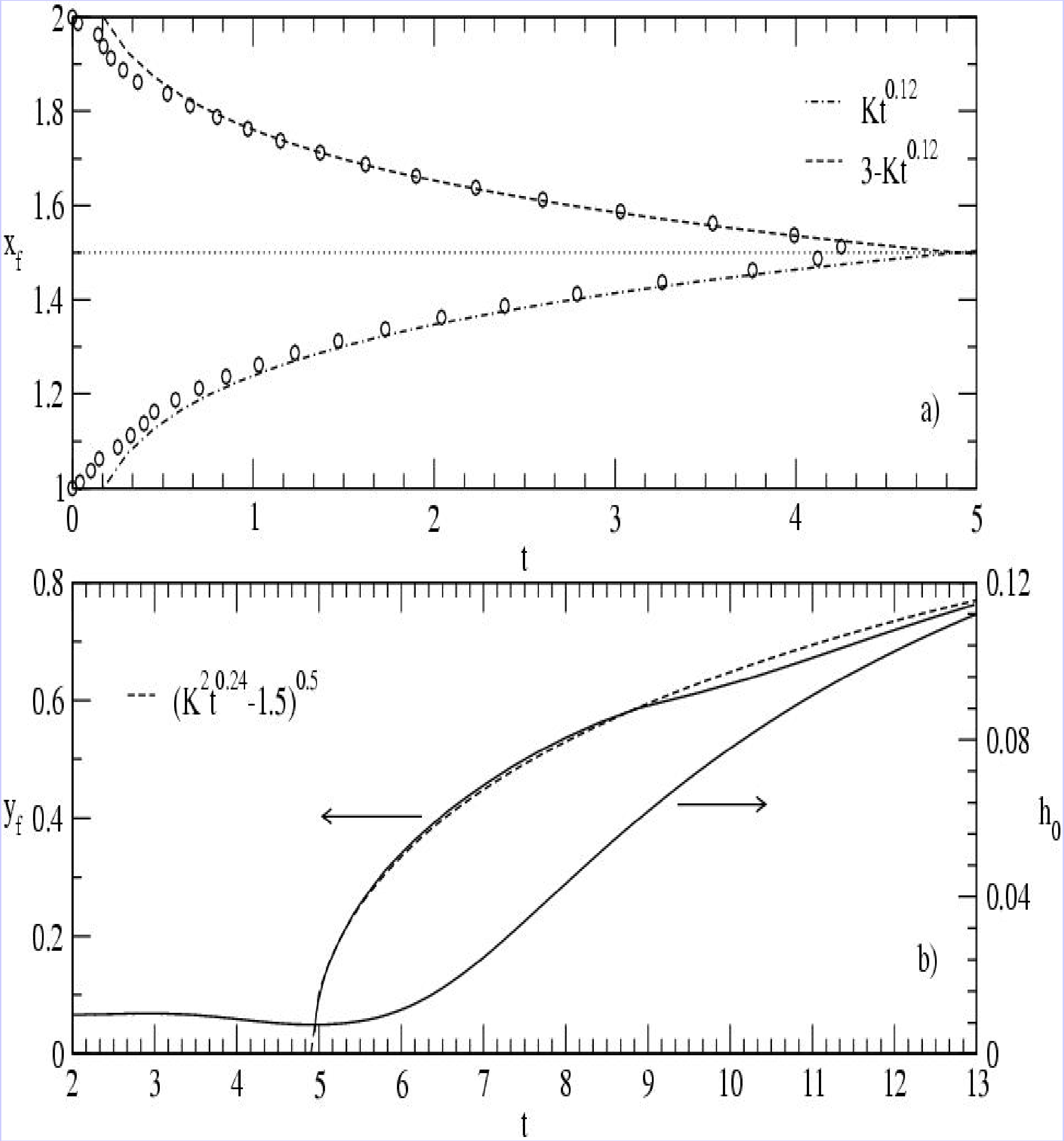


Coalescence: connection to theory

Numerics lets us compare to self-similar solutions

Extension to more drops possible
Diez & Kondic JCP '02

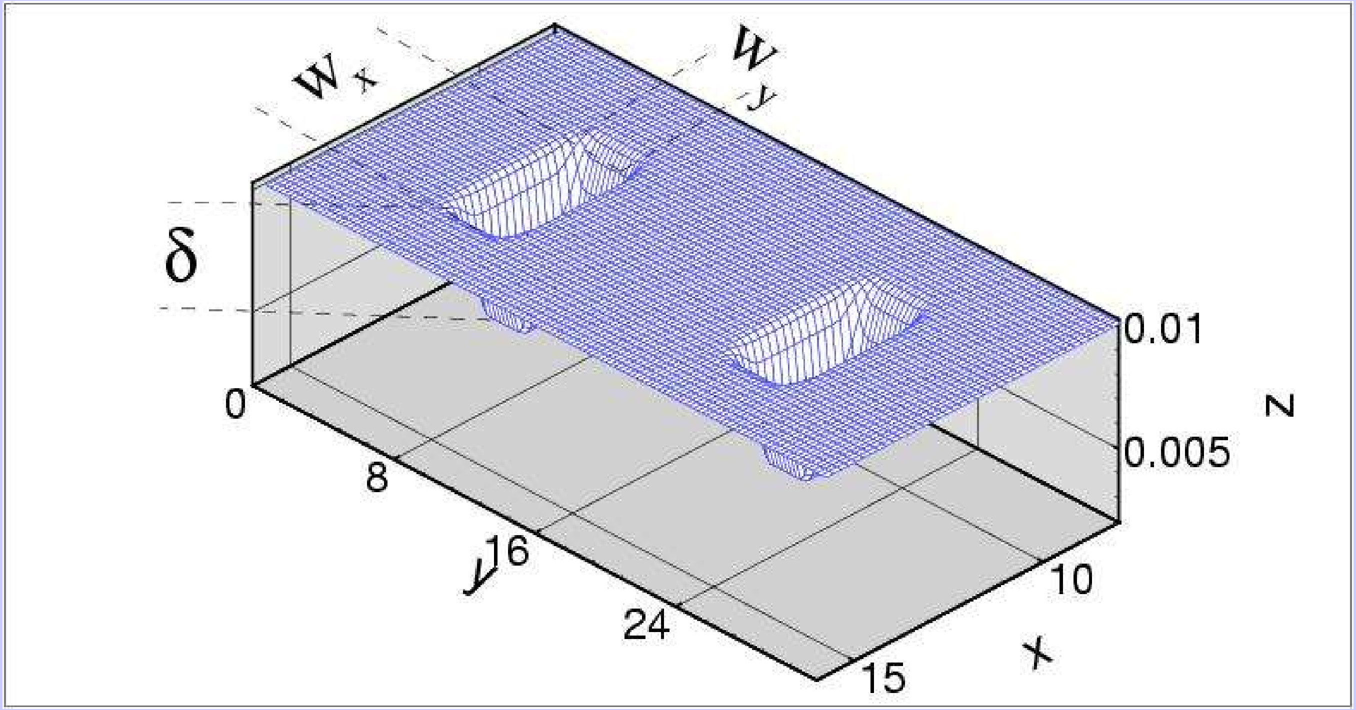




Application: Flow on Patterned Surfaces and Substrate Noise

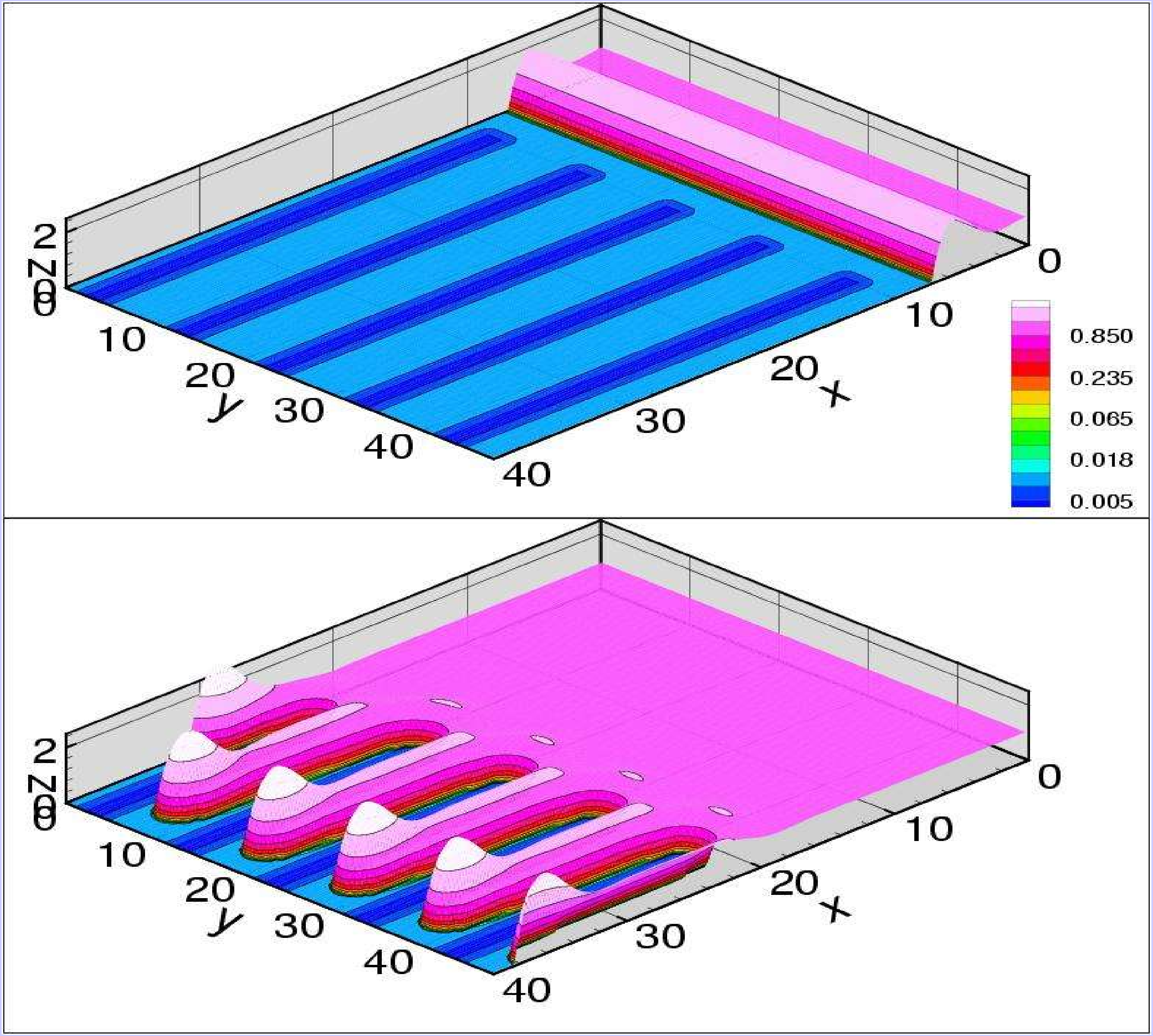
- Idea: modify surface properties in controllable manner and analyze how imposed surface features modify the flow
- Questions
  - Can one force the fluid to follow substrate features?
  - What is the interaction between imposed features and the natural fluid instability?
- Simple model: perturb precursor film thickness and analyze the flow

Precursor perturbation



What happens as the fluid flow over these perturbations?

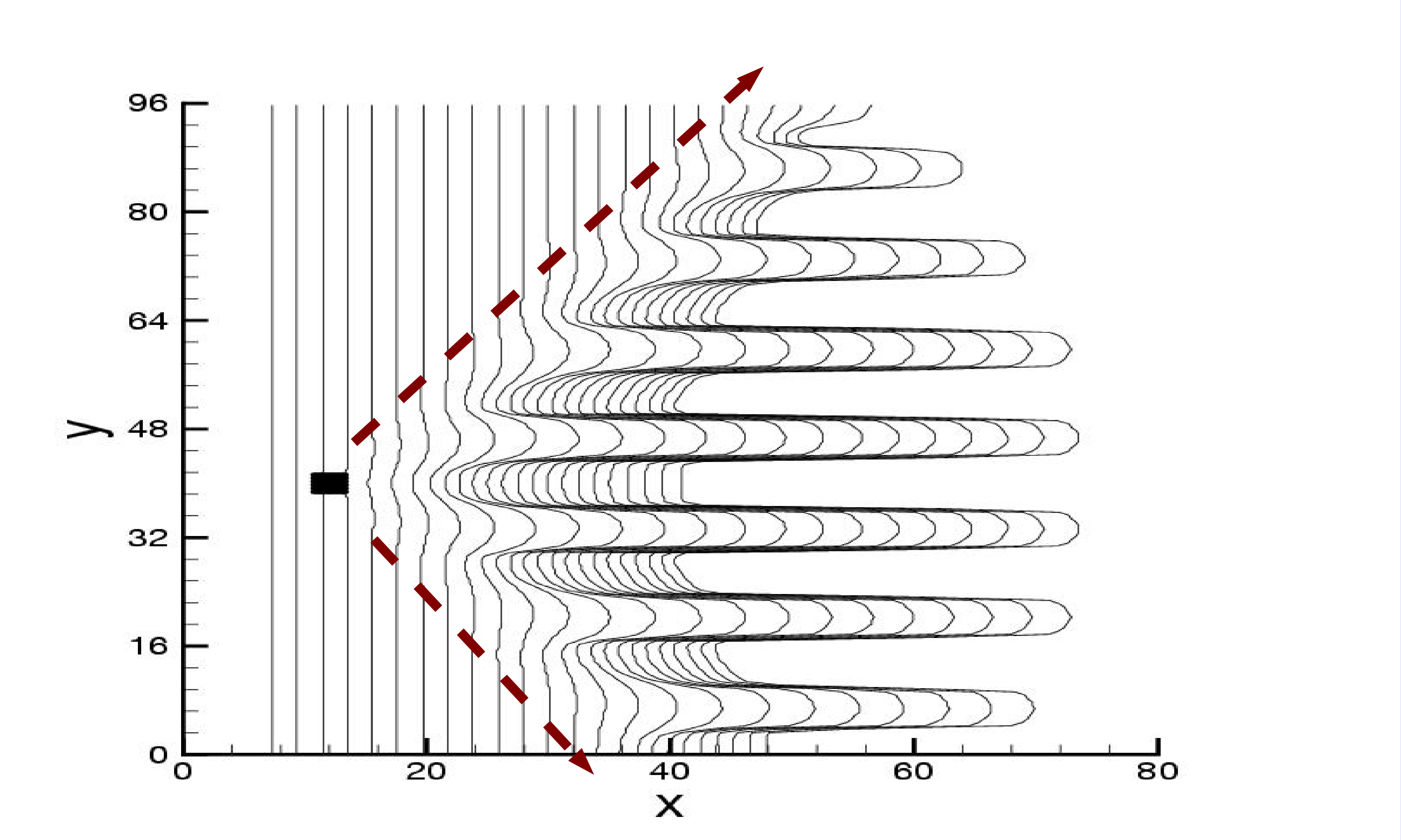
Result of imposed perturbations



Basic results

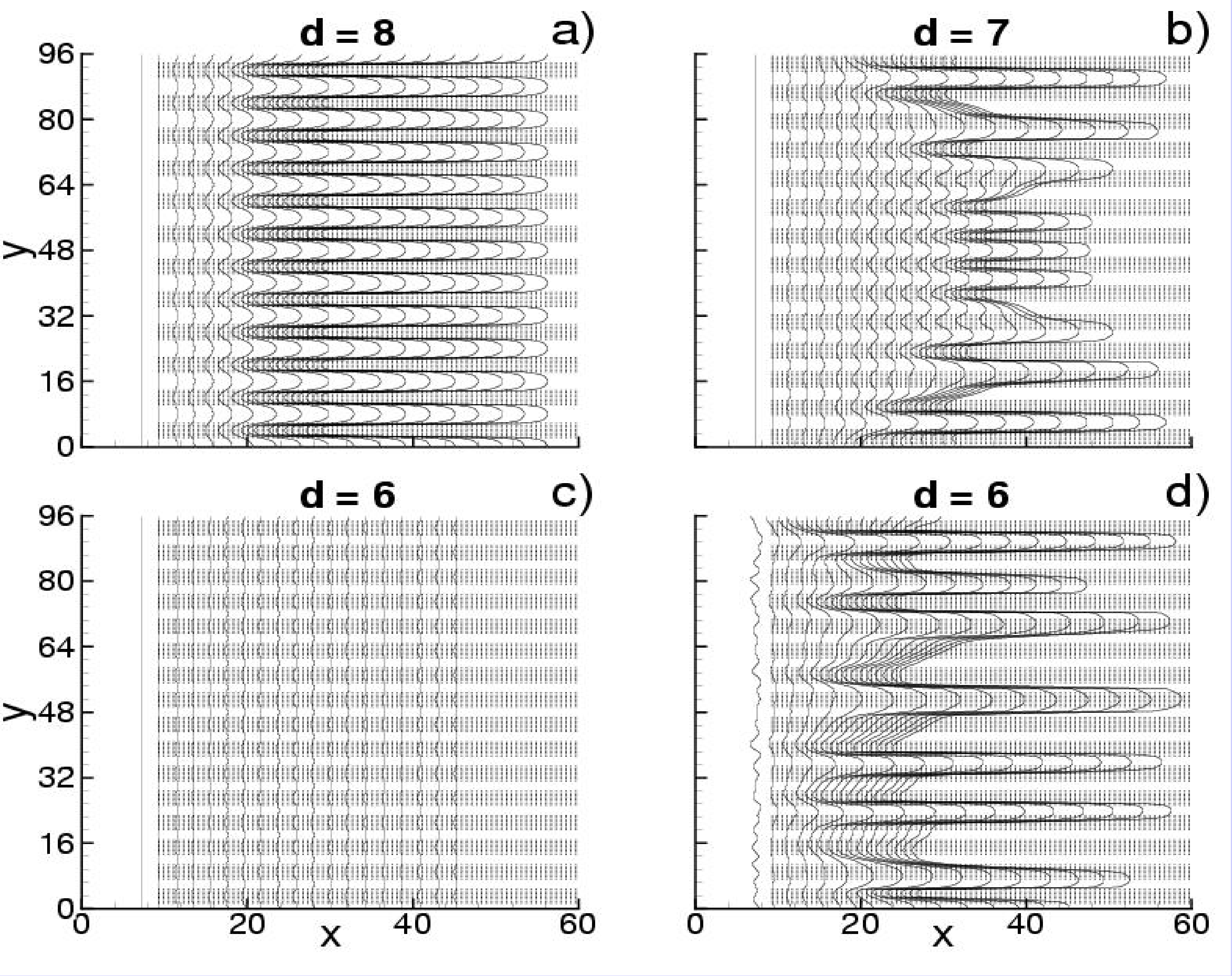
- Simulations show that instability could be imposed by the precursor perturbations
- The resulting patterns are similar to the previously obtained ones
  - Universality of the instability mechanism?
- Does the fluid always follow the imposed perturbations?
  - What is the influence of noise?
  - How fast does the information propagate through the fluid?

Information propagation



- Simple (approximate) answer follows from LSA: $v \approx \lambda^* \sigma^*$

Some more numerical experiments



What is numerics telling us?

- Fluid pattern cannot developed below linear stability limit
- Influence of noise is crucial
  - More details: Kondic & Diez, PRE '02, PoF '04
- Very similar results as in experiments where fluid wetting properties are perturbed
  - Troian, Nature '99

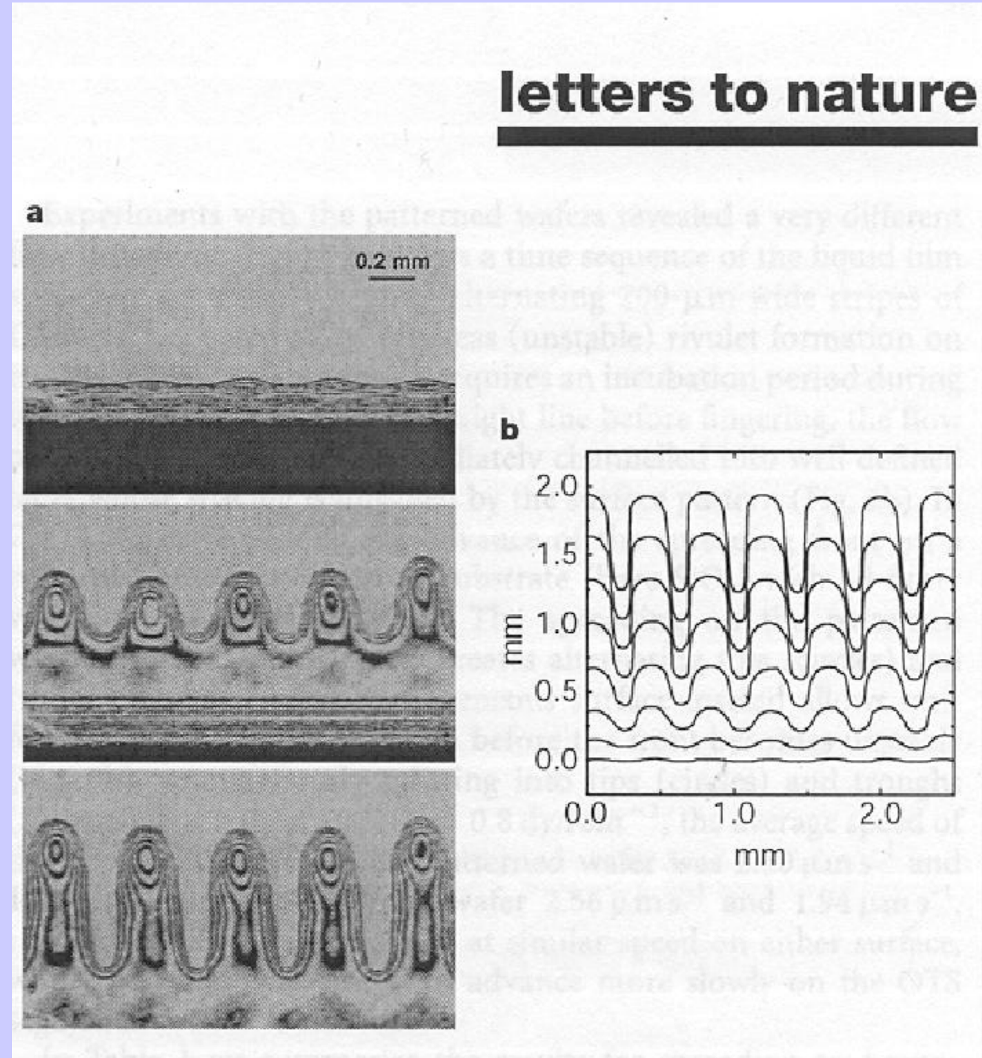


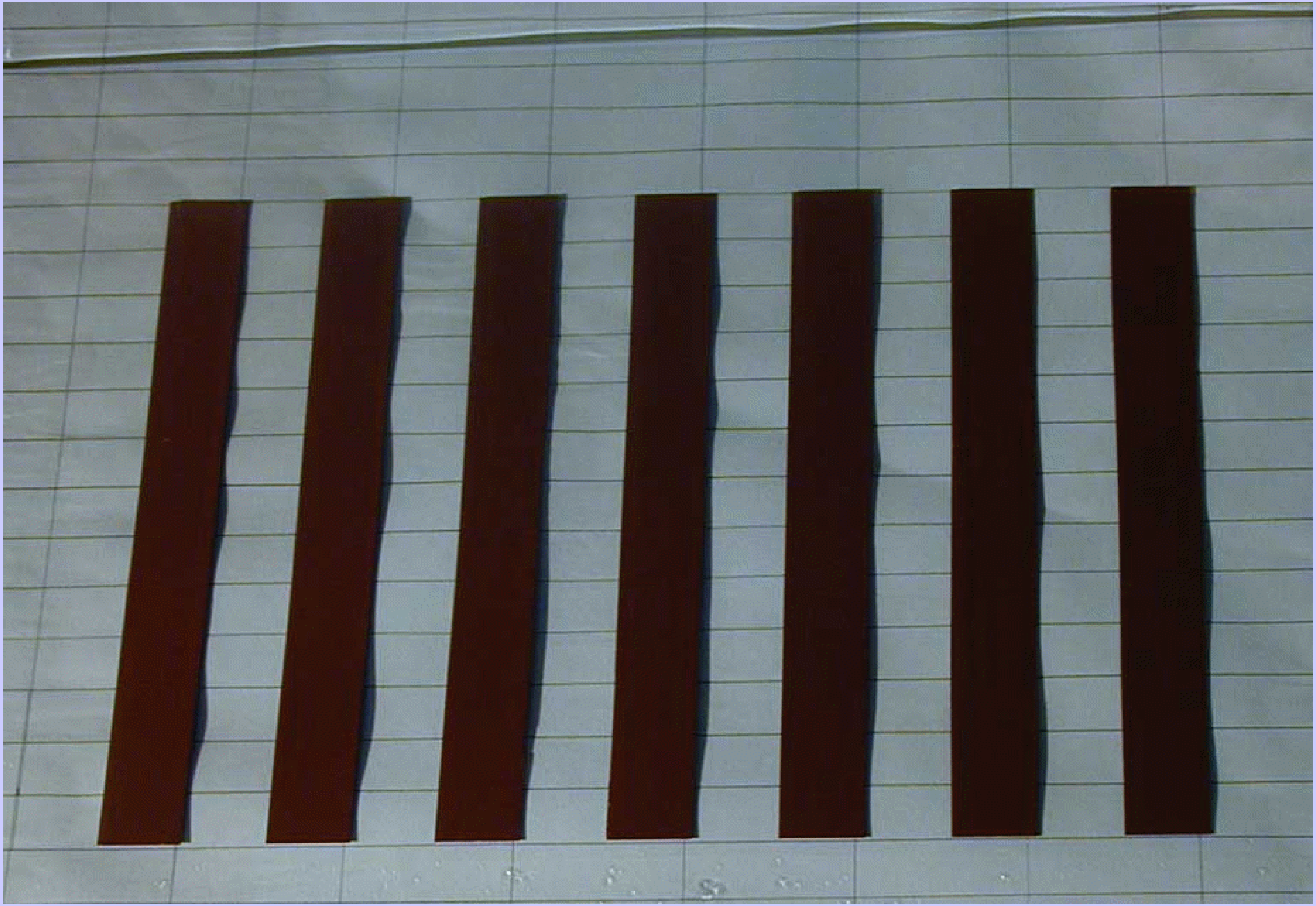
Figure 2 Time series of a silicone-oil film spreading on a patterned silicon wafer. The pattern consists of alternating 200-μm-wide stripes of OTS and bare oxidized SiO_2, and the spreading was monitored by interferometry. **a**, Three interferograms, with a five minute lapse between each. Adjacent fringes correspond to a change in film thickness of 0.2255 μm. **b**, A series of digitized curves, with a two minute lapse between each, representing the liquid front. The applied shear stress for **a** and **b** is $\tau = 0.8$ dyn cm^{-2}.

Experiments at undergraduate lab at NJIT

- Silicon oil on glass: good wetting
- Flow on unperturbed and on perturbed surfaces
- Excellent project to convince students that math actually works! (LSA + simple numerics could be compared to rather complex experimental results)
  - Kondic, SIAM Review '03

NEW YORK
GIANTS

Experiments on perturbed substrates



Application: Flow of Partially Wetting Fluids

- Flow of partially wetting fluids requires some more care in modeling
  - Mostly work by Javier Diez and his collaborators at Tandil (see the poster A227 for interesting experimental results)
- One approach is inclusion of van der Waals forces via disjoining pressure model
  - Physical concept: include the fact that molecular forces in the thin film are different from the bulk fluid
  - Model this effect by correcting Laplace-Young condition at the fluid – air interface

Disjoining pressure model

$$-\gamma\kappa \rightarrow -\gamma\kappa - \Pi(h)$$

$$\Pi(h) \sim \frac{1}{\hat{h}^n}\left[\left(\frac{\hat{h}}{h}\right)^n - \left(\frac{\hat{h}}{h}\right)^m\right]$$

$\hat{h}$: precursor film thickness

n, m: model dependent parameters

(e.g., n = 9, m = 3 result from Lennard-Jones potential)

Sliding drops

- Analyze how combination of fluid and flow parameters determines the dynamics

Small Sliding Drop

Large Sliding Drop

Two sliding Drops

More to come: work in progress

Next Steps

- Physics
  - Electrical effects
  - Evaporation
  - Thermal conduction
  - Complex fluids
  - Partial wetting
- Math/computing
  - Faster methods
  - Parallel computing
  - Theoretical methods
  - Extension to new geometries

- Applications: bridge the gap between `aca-demic' and `relevant' problems!